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10/576243
IAP15 Redacted by Inventor
(Name of person mailing paper or fee)Donna J. Vetter
Signature of person mailingHexylcarboxanilides

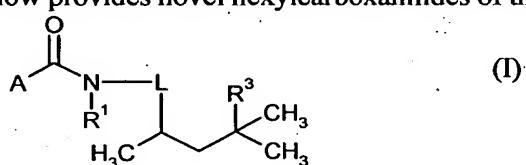
The present invention relates to novel hexylcarboxanilides, to a plurality of processes for their preparation and to their use for controlling unwanted microorganisms.

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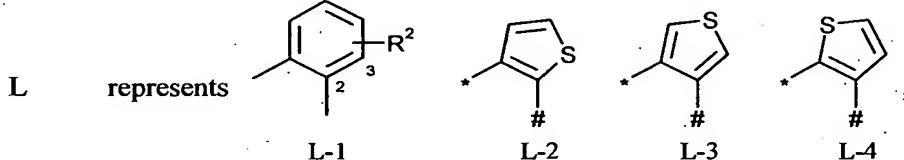
It is already known that numerous carboxanilides have fungicidal properties (cf., for example, WO 03/010149, WO 02/059086, WO 02/38542, WO 00/09482, EP-A 0 591 699, EP-A 0 589 301 and EP-A 0 545 099). Thus, for example, 5-fluoro-1,3-dimethyl-N-[2-(1,3,3-trimethylbutyl)phényl]-1H-pyrazole-4-carboxamide is known from WO 03/010149, N-allyl-N-[2-(1,3-dimethylbutyl)phenyl]-1-methyl-3-(trifluoromethyl)-1H-pyrazole-4-carboxamide is known from WO 02/059086 and N-[2-(1,3-dimethylbutyl)phenyl]-1-methyl-4-(trifluoromethyl)-1H-pyrrole-3-carboxamide is known from WO 02/38542. The activity of these compounds is good; however, at low application rates it is sometimes unsatisfactory.

10

15 This invention now provides novel hexylcarboxanilides of the formula (I)



in which



20

where the bond marked with * is attached to the amide, whereas the bond marked with # is attached to the alkyl side chain,

R¹

represents hydrogen, C₁-C₈-alkyl, C₁-C₆-alkylsulphanyl, C₁-C₆-alkylsulphonyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl; C₁-C₆-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄-haloalkylsulphanyl, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl; halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms;

25

(C₁-C₈-alkyl)carbonyl, (C₁-C₈-alkoxy)carbonyl, (C₁-C₄-alkoxy-C₁-C₄-alkyl)carbonyl, (C₃-C₈-cycloalkyl)carbonyl; (C₁-C₆-haloalkyl)carbonyl, (C₁-C₆-haloalkoxy)carbonyl, (halo-C₁-C₄-alkoxy-C₁-C₄-alkyl)carbonyl, (C₃-C₈-halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O)R⁴, -CONR⁵R⁶ or -CH₂NR⁷R⁸,

R²

represents hydrogen, fluorine, chlorine, methyl or trifluoromethyl,

R³ represents halogen, C₁-C₈-alkyl or C₁-C₈-haloalkyl,

R⁴ represents hydrogen, C₁-C₈-alkyl, C₁-C₈-alkoxy, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl, C₁-C₆-haloalkyl, C₁-C₆-haloalkoxy, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms,

5 R⁵ and R⁶ independently of one another each represent hydrogen, C₁-C₈-alkyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl; C₁-C₈-haloalkyl, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms,

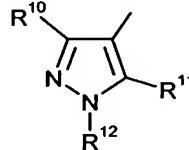
10 R⁵ and R⁶ furthermore together with the nitrogen atom to which they are attached form a saturated heterocycle having 5 to 8 ring atoms which is optionally mono- or polysubstituted by identical or different substituents from the group consisting of halogen and C₁-C₄-alkyl, where the heterocycle may contain 1 or 2 further non-adjacent heteroatoms from the group consisting of oxygen, sulphur and NR⁹,

15 R⁷ and R⁸ independently of one another represent hydrogen, C₁-C₈-alkyl, C₃-C₈-cycloalkyl; C₁-C₈-haloalkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms,

20 R⁷ and R⁸ furthermore together with the nitrogen atom to which they are attached form a saturated heterocycle having 5 to 8 ring atoms which is optionally mono- or polysubstituted by identical or different substituents from the group consisting of halogen and C₁-C₄-alkyl, where the heterocycle may contain 1 or 2 further non-adjacent heteroatoms from the group consisting of oxygen, sulphur and NR⁹,

R⁹ represents hydrogen or C₁-C₆-alkyl,

A represents the radical of the formula (A1)



(A1) in which

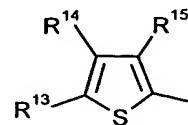
25 R¹⁰ represents hydrogen, hydroxyl, formyl, cyano, fluorine, chlorine, bromine, nitro, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₃-C₆-cycloalkyl, C₁-C₄-haloalkyl, C₁-C₄-haloalkoxy or C₁-C₄-haloalkylthio having in each case 1 to 5 halogen atoms, aminocarbonyl or aminocarbonyl-C₁-C₄-alkyl,

30 R¹¹ represents hydrogen, chlorine, bromine, iodine, cyano, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-haloalkyl or C₁-C₄-haloalkylthio having in each case 1 to 5 halogen atoms, and

35 R¹² represents hydrogen, C₁-C₄-alkyl, hydroxy-C₁-C₄-alkyl, C₂-C₆-alkenyl, C₃-C₆-cycloalkyl, C₁-C₄-alkylthio-C₁-C₄-alkyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio-C₁-C₄-alkyl, C₁-C₄-haloalkoxy-C₁-C₄-alkyl having in each case 1 to 5 halogen atoms, or represents phenyl,

or

A represents the radical of the formula (A2)



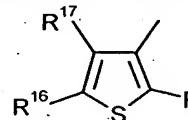
(A2) in which

5 R^{13} and R^{14} independently of one another represent hydrogen, halogen, C_1 - C_4 -alkyl or C_1 - C_4 -haloalkyl having in each case 1 to 5 halogen atoms and

R^{15} represents halogen, cyano or C_1 - C_4 -alkyl, or C_1 - C_4 -haloalkyl or C_1 - C_4 -haloalkoxy having in each case 1 to 5 halogen atoms,

or

A represents the radical of the formula (A3)



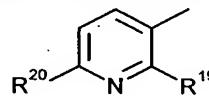
(A3) in which

10 R^{16} and R^{17} independently of one another represent hydrogen, halogen, C_1 - C_4 -alkyl or C_1 - C_4 -haloalkyl having 1 to 5 halogen atoms and

R^{18} represents hydrogen, C_1 - C_4 -alkyl or C_1 - C_4 -haloalkyl having 1 to 5 halogen atoms,

or

A represents the radical of the formula (A4)



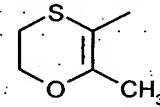
(A4) in which

15 R^{19} represents halogen, hydroxyl, cyano, C_1 - C_4 -alkyl, C_1 - C_4 -alkoxy, C_1 - C_4 -alkylthio, C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkylthio or C_1 - C_4 -haloalkoxy having in each case 1 to 5 halogen atoms and

R^{20} represents hydrogen, halogen, cyano, C_1 - C_4 -alkyl, C_1 - C_4 -alkoxy, C_1 - C_4 -alkylthio, C_1 - C_4 -haloalkyl, C_1 - C_4 -haloalkoxy having in each case 1 to 5 halogen atoms, C_1 - C_4 -alkylsulphinyl or C_1 - C_4 -alkylsulphonyl,

or

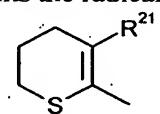
A represents the radical of the formula (A5)



(A5),

or

25 A represents the radical of the formula (A6)

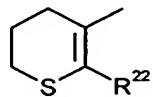


(A6) in which

R^{21} represents C_1 - C_4 -alkyl or C_1 - C_4 -haloalkyl having 1 to 5 halogen atoms,

or

A represents the radical of the formula (A7)

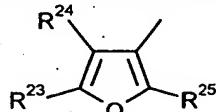


(A7) in which

15 R²² represents C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

20 A represents the radical of the formula (A8)



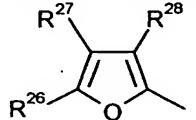
(A8) in which

5 R²³ and R²⁴ independently of one another represent hydrogen, halogen, amino, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms and

10 R²⁵ represents hydrogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

15 A represents the radical of the formula (A9)



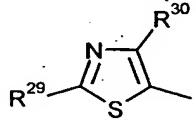
(A9) in which

15 R²⁶ and R²⁷ independently of one another represent hydrogen, halogen, amino, nitro, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms and

20 R²⁸ represents halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

25 A represents the radical of the formula (A10)



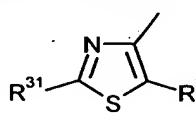
(A10) in which

20 R²⁹ represents hydrogen, halogen, amino, C₁-C₄-alkylamino, di-(C₁-C₄-alkyl)amino, cyano, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms and

25 R³⁰ represents halogen, hydroxyl, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₃-C₆-cycloalkyl, C₁-C₄-haloalkyl or C₁-C₄-haloalkoxy having in each case 1 to 5 halogen atoms,

or

30 A represents the radical of the formula (A11)



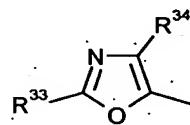
(A11) in which

25 R³¹ represents hydrogen, halogen, amino, C₁-C₄-alkylamino, di-(C₁-C₄-alkyl)amino, cyano, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms and

30 R³² represents halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

35 A represents the radical of the formula (A12)

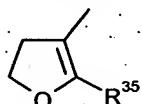


(A12) in which

R³³ represents hydrogen or C₁-C₄-alkyl andR³⁴ represents halogen or C₁-C₄-alkyl,

or

5 A represents the radical of the formula (A13)

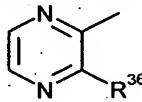


(A13) in which

R³⁵ represents C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

A represents the radical of the formula (A14)



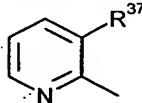
(A14) in which

10

R³⁶ represents hydrogen, halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

A represents the radical of the formula (A15)



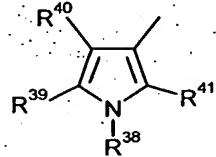
(A15) in which

15

R³⁷ represents halogen, hydroxyl, C₁-C₄-alkyl, C₁-C₄-alkoxy, C₁-C₄-alkylthio, C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio or C₁-C₄-haloalkoxy having in each case 1 to 5 halogen atoms;

or

20 A represents the radical of the formula (A16)



(A16) in which

25

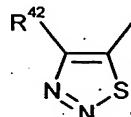
R³⁸ represents hydrogen, cyano, C₁-C₄-alkyl, C₁-C₄-haloalkyl having 1 to 5 halogen atoms, C₁-C₄-alkoxy-C₁-C₄-alkyl, hydroxy-C₁-C₄-alkyl, C₁-C₄-alkylsulphonyl, di(C₁-C₄-alkyl)aminosulphonyl, C₁-C₆-alkylcarbonyl or in each case optionally substituted phenylsulphonyl or benzoyl,R³⁹ represents hydrogen, halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,R⁴⁰ represents hydrogen, halogen, cyano, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

5 R^{41} represents hydrogen, halogen, C₁-C₄-alkyl or C₁-C₄-haloalkyl having 1 to 5 halogen atoms,

or

10 A represents the radical of the formula (A17)

5



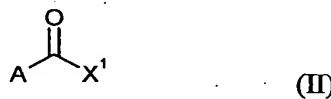
(A17) in which

15 R^{42} represents C₁-C₄-alkyl.

The compounds according to the invention can, if appropriate, be present as mixtures of various possible isomeric forms, in particular of stereoisomers, such as, for example, E and Z, threo and erythro and also optical isomers, and, if appropriate, also of tautomers. What is claimed are both the E and the Z isomers, and also the threo and erythro and the optical isomers, any mixtures of these isomers and the possible tautomeric forms.

15 Furthermore, it has been found that hexylcarboxanilides of the formula (I) are obtained when

a) carboxylic acid derivatives of the formula (II)

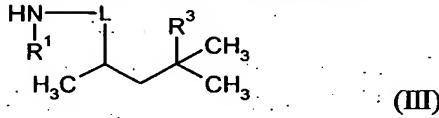


in which

20 A is as defined above and

X^1 represents halogen or hydroxyl

are reacted with an aniline derivative of the formula (III)

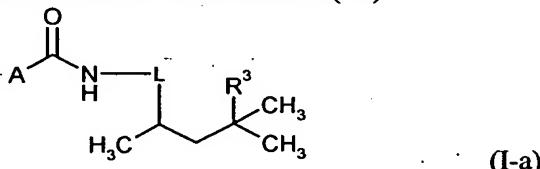


in which L, R¹ and R³ are as defined above,

25 if appropriate in the presence of a catalyst, if appropriate in the presence of a condensing agent, if appropriate in the presence of an acid binder and if appropriate in the presence of a diluent,

or

30 b) hexylcarboxanilides of the formula (I-a)



in which L, A and R³ are as defined above

are reacted with halides of the formula (IV)



in which

X^2 represents chlorine, bromine or iodine,

5 R^{1-A} represents C_1-C_8 -alkyl, C_1-C_6 -alkylsulphonyl, C_1-C_6 -alkylsulphonyl, C_1-C_4 -alkoxy- C_1-C_4 -alkyl, C_3-C_8 -cycloalkyl; C_1-C_6 -haloalkyl, C_1-C_4 -haloalkylthio, C_1-C_4 -haloalkylsulphonyl, C_1-C_4 -haloalkylsulphonyl, halo- C_1-C_4 -alkoxy- C_1-C_4 -alkyl, C_3-C_8 -halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl- C_1-C_3 -alkyl, (C_1-C_3 -alkyl)carbonyl- C_1-C_3 -alkyl, (C_1-C_3 -alkoxy)carbonyl- C_1-C_3 -alkyl; halo-(C_1-C_3 -alkyl)carbonyl- C_1-C_3 -alkyl, halo-(C_1-C_3 -alkoxy)carbonyl- C_1-C_3 -alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms; (C_1-C_8 -alkyl)carbonyl, (C_1-C_8 -alkoxy)carbonyl, (C_1-C_4 -alkoxy- C_1-C_4 -alkyl)carbonyl, (C_3-C_8 -cycloalkyl)carbonyl; (C_1-C_6 -haloalkyl)carbonyl, (C_1-C_6 -haloalkoxy)carbonyl, (halo- C_1-C_4 -alkoxy- C_1-C_4 -alkyl)carbonyl, (C_3-C_8 -halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or $-C(=O)C(=O)R^4$, $-CONR^5R^6$ or $-CH_2NR^7R^8$,

where R^4 , R^5 , R^6 , R^7 and R^8 are as defined above

in the presence of a base and in the presence of a diluent.

20

Finally, it has been found that the novel hexylcarboxanilides of the formula (I) have very good microbicidal properties and can be used for controlling unwanted microorganisms both in crop protection and in the protection of materials.

25

The formula (I) provides a general definition of the hexylcarboxanilides according to the invention. Preferred radical definitions of the formulae shown above and below are given below. These definitions apply both to the end products of the formula (I) and likewise to all intermediates.

30 L preferably represents $L-1$ where R^2 may in each case have the general, preferred, particularly preferred, very particularly preferred or especially preferred meanings.

L furthermore preferably represents $L-2$.

L furthermore preferably represents $L-3$.

L furthermore preferably represents $L-4$.

35 L particularly preferably represents $L-1$, where R^2 may in each case have the general, preferred, particularly preferred, very particularly preferred or especially preferred meanings.

L furthermore particularly preferably represents $L-2$.

L very particularly preferably represents $L-1$ where R^2 may in each case have the general, preferred, particularly preferred, very particularly preferred or especially preferred meanings.

5 R¹ preferably represents hydrogen, C₁-C₆-alkyl, C₁-C₄-alkylsulphanyl, C₁-C₄-alkylsulphonyl, C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-cycloalkyl; C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄-haloalkylsulphanyl, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl; halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms; (C₁-C₆-alkyl)carbonyl, (C₁-C₄-alkoxy)carbonyl, (C₁-C₃-alkoxy-C₁-C₃-alkyl)carbonyl, (C₃-C₆-cycloalkyl)carbonyl; (C₁-C₄-haloalkyl)carbonyl, (C₁-C₄-haloalkoxy)carbonyl, (halo-C₁-C₃-alkoxy-C₁-C₃-alkyl)carbonyl, (C₃-C₆-halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O)R⁴, -CONR⁵R⁶ or -CH₂NR⁷R⁸.

10 R¹ particularly preferably represents hydrogen, methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl, pentyl or hexyl, methylsulphanyl, ethylsulphanyl, n- or isopropylsulphanyl, n-, iso-, sec- or tert-butylsulphanyl, methylsulphonyl, ethylsulphonyl, n- or isopropylsulphonyl, n-, iso-, sec- or tert-butylsulphonyl, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, cyclopropyl, cyclopentyl, cyclohexyl, trifluoromethyl, trichloromethyl, trifluoroethyl, difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethylsulphanyl, trifluoromethylsulphonyl, trifluoromethoxymethyl; formyl, -CH₂-CHO, -(CH₂)₂-CHO, -CH₂-CO-CH₃, -CH₂-CO-CH₂CH₃, -CH₂-CO-CH(CH₃)₂, -(CH₂)₂-CO-CH₃, -(CH₂)₂-CO-CH₂CH₃, -CH₂-CO₂CH₂CH₃, -CH₂-CO₂CH(CH₃)₂, -CH₂-CO-CF₃, -CH₂-CO-CCl₃, -CH₂-CO-CH₂CF₃, -CH₂-CO-CH₂CCl₃, -(CH₂)₂-CO-CH₂CCl₃, -CH₂-CO₂CH₂CF₃, -(CH₂)₂-CO₂CF₂CF₃, -CH₂-CO₂CH₂CCl₃, -CH₂-CO₂CCl₂CCl₃, -(CH₂)₂-CO₂CH₂CF₃, -(CH₂)₂-CO₂CF₂CF₃, -(CH₂)₂-CO₂CH₂CCl₃, -(CH₂)₂-CO₂CCl₂CCl₃;

15 methylcarbonyl, ethylcarbonyl, n-propylcarbonyl, isopropylcarbonyl, tert-butylcarbonyl, methoxycarbonyl, ethoxycarbonyl, tert-butoxycarbonyl, cyclopropylcarbonyl; trifluoromethylcarbonyl, trifluoromethoxycarbonyl, or -C(=O)C(=O)R⁵, -CONR⁶R⁷ or -CH₂NR⁸R⁹.

20 R¹ very particularly preferably represents hydrogen, methyl, methoxymethyl, formyl, -CH₂-CHO, -(CH₂)₂-CHO, -CH₂-CO-CH₃, -CH₂-CO-CH₂CH₃, -CH₂-CO-CH(CH₃)₂, -C(=O)CHO, -C(=O)C(=O)CH₃, -C(=O)C(=O)CH₂OCH₃, -C(=O)CO₂CH₃, -C(=O)CO₂CH₂CH₃.

25 R² preferably represents hydrogen.

30 R² furthermore preferably represents fluorine, where fluorine is particularly preferably located in the 4-, 5- or 6-position, very particularly preferably in the 4- or 6-position, especially in the 4-position, of the anilide radical [cf. formula (I) above].

R² furthermore preferably represents chlorine, where chlorine is particularly preferably located in the 5-position of the anilide radical [cf. formula (I) above]. Chlorine is furthermore particularly preferably located in the 4-position of the anilide radical.

5 R² furthermore preferably represents methyl, where methyl is particularly preferably located in the 3-position of the anilide radical [cf. formula (I) above].

R² furthermore preferably represents trifluoromethyl, where trifluoromethyl is particularly preferably located in the 4- or 5-position of the anilide radical [cf. formula (I) above].

10 R³ preferably represents fluorine, chlorine, bromine, iodine, C₁-C₆-alkyl, C₁-C₆-haloalkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms.

R³ particularly preferably represents fluorine, chlorine, bromine, methyl, ethyl, n-, isopropyl, n-, iso-, sec-, tert-butyl or C₁-C₄-haloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms.

15 R³ very particularly preferably represents fluorine, chlorine, methyl, ethyl or trifluoromethyl.

R⁴ preferably represents hydrogen, C₁-C₆-alkyl, C₁-C₄-alkoxy, C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-cycloalkyl; C₁-C₄-haloalkyl, C₁-C₄-haloalkoxy, halo-C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms.

20 R⁴ particularly preferably represents hydrogen, methyl, ethyl, n- or isopropyl, tert-butyl, methoxy, ethoxy, n- or isopropoxy, tert-butoxy, methoxymethyl, cyclopropyl; trifluoromethyl, trifluoromethoxy.

25 R⁵ and R⁶ independently of one another preferably represent hydrogen, C₁-C₆-alkyl, C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-cycloalkyl; C₁-C₄-haloalkyl, halo-C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₆-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms.

30 R⁵ and R⁶ furthermore together with the nitrogen atom to which they are attached preferably represent a saturated heterocycle having 5 or 6 ring atoms which is optionally mono- to tetrasubstituted by identical or different substituents from the group consisting of halogen and C₁-C₄-alkyl, where the heterocycle may contain 1 or 2 further non-adjacent heteroatoms from the group consisting of oxygen, sulphur and NR⁹.

35 R⁵ and R⁶ independently of one another particularly preferably represent hydrogen, methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, cyclopropyl, cyclopentyl, cyclohexyl; trifluoromethyl, trichloromethyl, trifluoroethyl, trifluoromethoxymethyl.

R⁵ and R⁶ furthermore together with the nitrogen atom to which they are attached particularly preferably represent a saturated heterocycle from the group consisting of morpholine, thiomorpholine and piperazine which is optionally mono- to tetrasubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine and methyl, where the piperazine may be substituted on the second nitrogen atom by R⁹.

R^7 and R^8 independently of one another preferably represent hydrogen, C_1 - C_6 -alkyl, C_3 - C_6 -cycloalkyl; C_1 - C_4 -haloalkyl, C_3 - C_6 -halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms.

5 R^7 and R^8 furthermore together with the nitrogen atom to which they are attached preferably represent a saturated heterocycle having 5 or 6 ring atoms which is optionally mono- or polysubstituted by identical or different substituents from the group consisting of halogen and C_1 - C_4 -alkyl, where the heterocycle may contain 1 or 2 further non-adjacent heteroatoms from the group consisting of oxygen, sulphur and NR^9 .

10 R^7 and R^8 independently of one another particularly preferably represent hydrogen, methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, cyclopropyl, cyclopentyl, cyclohexyl; trifluoromethyl, trichloromethyl, trifluoroethyl, trifluoromethoxymethyl.

15 R^7 and R^8 furthermore together with the nitrogen atom to which they are attached particularly preferably represent a saturated heterocycle from the group consisting of morpholine, thiomorpholine and piperazine which is optionally mono- to tetrasubstituted by identical or different substituents from the group consisting of fluorine, chlorine, bromine and methyl, where the piperazine may be substituted on the second nitrogen atom by R^9 .

20 R^9 preferably represents hydrogen or C_1 - C_4 -alkyl.
 R^9 particularly preferably represents hydrogen, methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl.

25 A particularly represents one of the radicals
 A1, A2, A3, A4, A5, A8, A9, A10, A11, A13, A15, A16 or A17 given above.

30 A particularly preferably represents one of the radicals
 A1, A2, A4, A5, A8, A10, A11, A13, A15, A16 or A17 given above.
 A very particularly preferably represents the radical A1.
 A furthermore very particularly preferably represents the radical A2.
 A furthermore very particularly preferably represents the radical A4.
 A furthermore very particularly preferably represents the radical A5.
 A furthermore very particularly preferably represents the radical A8.
 A furthermore very particularly preferably represents the radical A10.
 A furthermore very particularly preferably represents the radical A11.
 A furthermore very particularly preferably represents the radical A13.
 A furthermore very particularly preferably represents the radical A15.
 A furthermore very particularly preferably represents the radical A17.

5 R¹⁰ preferably represents hydrogen, hydroxyl, formyl, cyano, fluorine, chlorine, bromine, methyl, ethyl, isopropyl, methoxy, ethoxy, methylthio, ethylthio, cyclopropyl, C₁-C₂-haloalkyl, C₁-C₂-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms, trifluoromethylthio, difluoromethylthio, aminocarbonyl, aminocarbonylmethyl or aminocarbonylethyl.

10 R¹⁰ particularly preferably represents hydrogen, hydroxyl, formyl, fluorine, chlorine, bromine, methyl, ethyl, isopropyl, methoxy, ethoxy, monofluoromethyl, monofluoroethyl, difluoromethyl, trifluoromethyl, difluorochloromethyl, trichloromethyl, dichloromethyl, pentafluoroethyl, cyclopropyl, methoxy, ethoxy, trifluoromethoxy, difluoromethoxy, trichloromethoxy, methylthio, ethylthio, trifluoromethylthio or difluoromethylthio.

15 R¹⁰ very particularly preferably represents hydrogen, hydroxyl, formyl, fluorine, chlorine, bromine, methyl, ethyl, isopropyl, methoxy, cyclopropyl, monofluoromethyl, monofluoroethyl, difluoromethyl, dichloromethyl, trifluoromethyl, difluorochloromethyl, trichloromethyl, -CHFCH₃ or difluoromethoxy.

20 R¹⁰ especially preferably represents hydrogen, hydroxyl, formyl, chlorine, methyl, ethyl, methoxy, cyclopropyl, monofluoromethyl, difluoromethyl, dichloromethyl, trifluoromethyl, -CHFCH₃ or difluoromethoxy.

25 R¹¹ preferably represents hydrogen, chlorine, bromine, iodine, methyl, ethyl, methoxy, ethoxy, methylthio, ethylthio, C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms,

R¹¹ particularly preferably represents hydrogen, chlorine, bromine, iodine, methyl or -CHFCH₃.

R¹¹ very particularly preferably represents hydrogen, chlorine, methyl or -CHFCH₃.

30 R¹² preferably represents hydrogen, methyl, ethyl, n-propyl, isopropyl, C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms, hydroxymethyl, hydroxyethyl, cyclopropyl, cyclopentyl, cyclohexyl or phenyl.

R¹² particularly preferably represents hydrogen, methyl, ethyl, isopropyl, trifluoromethyl, difluoromethyl, hydroxymethyl, hydroxyethyl or phenyl.

R¹² very particularly preferably represents hydrogen, methyl, trifluoromethyl or phenyl.

35 R¹² especially preferably represents methyl.

R¹³ and R¹⁴ independently of one another preferably represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

40 R¹³ and R¹⁴ independently of one another particularly preferably represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, difluoromethyl, trifluoromethyl, difluorochloromethyl or trichloromethyl.

R¹³ and R¹⁴ independently of one another very particularly preferably represent hydrogen, fluorine, chlorine, bromine or methyl.

R¹³ and R¹⁴ especially preferably each represent hydrogen.

15 R^{15} preferably represents fluorine, chlorine, bromine, iodine, cyano, methyl, ethyl, C_1-C_2 -haloalkyl or C_1-C_2 -haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.

5 R^{15} particularly preferably represents fluorine, chlorine, bromine, iodine, cyano, methyl, trifluoromethyl, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy or trichloromethoxy.

R^{15} very particularly preferably represents fluorine, chlorine, bromine, iodine, methyl, trifluoromethyl or trifluoromethoxy.

10 R^{15} especially preferably represents chlorine or methyl.

R^{16} and R^{17} independently of one another preferably represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C_1-C_2 -haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

R^{16} and R^{17} independently of one another particularly preferably represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, difluoromethyl, trifluoromethyl, difluorochloromethyl or trichloromethyl.

15 R^{16} and R^{17} independently of one another very particularly preferably represent hydrogen, fluorine, chlorine, bromine or methyl.

R^{16} and R^{17} especially preferably each represent hydrogen.

20 R^{18} preferably represents hydrogen, methyl, ethyl or C_1-C_2 -haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

R^{18} particularly preferably represents hydrogen, methyl or trifluoromethyl.

R^{18} very particularly preferably represents methyl.

25 R^{19} preferably represents fluorine, chlorine, bromine, iodine, hydroxyl, cyano, C_1-C_4 -alkyl, methoxy, ethoxy, methylthio, ethylthio, difluoromethylthio, trifluoromethylthio, C_1-C_2 -haloalkyl or C_1-C_2 -haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.

30 R^{19} particularly preferably represents fluorine, chlorine, bromine, iodine, hydroxyl, cyano, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, trifluoromethyl, difluoromethyl, difluorochloromethyl, trichloromethyl, methoxy, ethoxy, methylthio, ethylthio, difluoromethylthio, trifluoromethylthio, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy or trichloromethoxy.

35 R^{19} very particularly preferably represents fluorine, chlorine, bromine, iodine, hydroxyl, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

R^{20} preferably represents hydrogen, fluorine, chlorine, bromine, iodine, cyano, C_1-C_4 -alkyl, methoxy, ethoxy, methylthio, ethylthio, C_1-C_2 -haloalkyl or C_1-C_2 -haloalkoxy having in each

case 1 to 5 fluorine, chlorine and/or bromine atoms, C₁-C₂-alkylsulphanyl or C₁-C₂-alkylsulphonyl.

5 R²⁰ particularly preferably represents hydrogen, fluorine, chlorine, bromine, iodine, cyano, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, trifluoromethyl, difluoromethyl, difluorochloromethyl, trichloromethyl, methoxy, ethoxy, methylthio, ethylthio, trifluoromethoxy, difluoromethoxy, difluorochloromethoxy, trichloromethoxy, methylsulphanyl or methylsulphonyl.

10 R²⁰ very particularly preferably represents hydrogen, fluorine, chlorine, bromine, iodine, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, trifluoromethyl, difluoromethyl, trichloromethyl, methylsulphanyl or methylsulphonyl.

15 R²⁰ especially preferably represents hydrogen or trifluoromethyl.

R²¹ preferably represents methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

20 R²¹ particularly preferably represents methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

R²¹ very particularly preferably represents methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

25 R²² preferably represents methyl, ethyl trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

R²² particularly preferably represents methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

30 R²³ and R²⁴ independently of one another preferably represent hydrogen, fluorine, chlorine, bromine, amino, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

R²³ and R²⁴ independently of one another particularly preferably represent hydrogen, fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

35 R²³ and R²⁴ independently of one another very particularly preferably represent hydrogen, fluorine, chlorine, bromine or methyl.

R²³ and R²⁴ especially preferably each represent hydrogen.

R²⁵ preferably represents hydrogen, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

R²⁵ particularly preferably represents hydrogen, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

R²⁵ very particularly preferably represents hydrogen, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

R²⁵ especially preferably represents methyl or trifluoromethyl.

R²⁶ and R²⁷ independently of one another preferably represent hydrogen, fluorine, chlorine, bromine, amino, nitro, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

5 R²⁶ and R²⁷ independently of one another particularly preferably represent hydrogen, fluorine, chlorine, bromine, nitro, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

10 R²⁶ and R²⁷ independently of one another very particularly preferably represent hydrogen, fluorine, chlorine, bromine or methyl.

R²⁶ and R²⁷ especially preferably each represent hydrogen.

15 R²⁸ preferably represents fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

R²⁸ particularly preferably represents fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

R²⁸ very particularly preferably represents fluorine, chlorine, bromine, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

20 R²⁸ especially preferably represents methyl.

R²⁹ preferably represents hydrogen, fluorine, chlorine, bromine, amino, C₁-C₄-alkylamino, di(C₁-C₄-alkyl)amino, cyano, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

25 R²⁹ particularly preferably represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, cyano, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

R²⁹ very particularly preferably represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

30 R²⁹ especially preferably represents hydrogen, chlorine, amino, methylamino, dimethylamino, methyl or trifluoromethyl.

R³⁰ preferably represents fluorine, chlorine, bromine, hydroxyl, methyl, ethyl, methoxy, ethoxy, cyclopropyl, C₁-C₂-haloalkyl or C₁-C₂-haloalkoxy having 1 to 5 fluorine, chlorine and/or bromine atoms.

35 R³⁰ particularly preferably represents fluorine, chlorine, bromine, hydroxyl, methyl, ethyl, methoxy, ethoxy, cyclopropyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

R³⁰ very particularly preferably represents, fluorine, chlorine, bromine, hydroxyl, methyl, methoxy, cyclopropyl, trifluoromethyl, difluoromethyl or trichloromethyl.

15 R^{31} preferably represents hydrogen, fluorine, chlorine, bromine, amino, C_1 - C_4 -alkylamino, di(C_1 - C_4 -alkyl)amino, cyano, methyl, ethyl or C_1 - C_2 -haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

5 R^{31} particularly preferably represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, cyano, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

10 R^{31} very particularly preferably represents hydrogen, fluorine, chlorine, bromine, amino, methylamino, dimethylamino, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

15 R^{31} especially preferably represents amino, methylamino, dimethylamino, methyl or trifluoromethyl.

20 R^{32} preferably represents fluorine, chlorine, bromine, methyl, ethyl or C_1 - C_2 -haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

15 R^{32} particularly preferably represents fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

25 R^{32} very particularly preferably represents fluorine, chlorine, bromine, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

20 R^{32} especially preferably represents methyl, trifluoromethyl or difluoromethyl.

25 R^{33} preferably represents hydrogen, methyl or ethyl.

25 R^{33} particularly preferably represents methyl.

25 R^{34} preferably represents fluorine, chlorine, bromine, methyl or ethyl.

25 R^{34} particularly preferably represents fluorine, chlorine or methyl.

30 R^{35} preferably represents methyl, ethyl or C_1 - C_2 -haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

30 R^{35} particularly preferably represents methyl, ethyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

35 R^{35} very particularly preferably represents methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

35 R^{35} especially preferably represents methyl or trifluoromethyl.

35 R^{36} preferably represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C_1 - C_2 -haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

35 R^{36} particularly preferably represents hydrogen, fluorine, chlorine, bromine, methyl or trifluoromethyl.

35 R^{36} very particularly preferably represents hydrogen or chlorine.

10 R³⁷ preferably represents fluorine, chlorine, bromine, iodine, hydroxyl, C₁-C₄-alkyl, methoxy, ethoxy, methylthio, ethylthio, difluoromethylthio, trifluoromethylthio, C₁-C₂-haloalkyl or C₁-C₂-haloalkoxy having in each case 1 to 5 fluorine, chlorine and/or bromine atoms.

15 R³⁷ particularly preferably represents fluorine, chlorine, bromine, iodine, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, sec-butyl, tert-butyl, trifluoromethyl, difluoromethyl, difluorochloromethyl, trichloromethyl.

20 R³⁷ very particularly preferably represents fluorine, chlorine, bromine, iodine, methyl, trifluoromethyl, difluoromethyl or trichloromethyl.

25 R³⁸ preferably represents hydrogen, methyl, ethyl, C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms, C₁-C₂-alkoxy-C₁-C₂-alkyl, hydroxymethyl, hydroxyethyl, methylsulphonyl or dimethylaminosulphonyl.

30 R³⁸ particularly preferably represents hydrogen, methyl, ethyl, trifluoromethyl, methoxymethyl, ethoxymethyl, hydroxymethyl or hydroxyethyl.

35 R³⁸ very particularly preferably represents methyl or methoxymethyl.

40 R³⁹ preferably represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

45 R³⁹ particularly preferably represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl, trifluoromethyl, difluoromethyl or trichloromethyl.

50 R³⁹ very particularly preferably represents hydrogen or methyl.

55 R⁴⁰ preferably represents hydrogen, fluorine, chlorine, bromine, cyano, methyl, ethyl, isopropyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

60 R⁴⁰ particularly preferably represents hydrogen, fluorine, chlorine, bromine, cyano, methyl, ethyl, isopropyl, trifluoromethyl, difluoromethyl, difluorochloromethyl or trichloromethyl.

65 R⁴⁰ very particularly preferably represents hydrogen, fluorine, methyl or trifluoromethyl.

70 R⁴¹ preferably represents hydrogen, fluorine, chlorine, bromine, methyl, ethyl or C₁-C₂-haloalkyl having 1 to 5 fluorine, chlorine and/or bromine atoms.

75 R⁴¹ particularly preferably represents hydrogen, fluorine, chlorine, bromine, iodine, methyl or trifluoromethyl.

80 R⁴¹ very particularly preferably represents hydrogen or trifluoromethyl.

85 R⁴² preferably represents methyl, ethyl, n-propyl or isopropyl.

90 R⁴² particularly preferably represents methyl or ethyl.

Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the general meanings given above.

Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the preferred meanings given above.

5 Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the particularly preferred meanings given above.

Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the very particularly preferred meanings given above.

10 Emphasis is given to compounds of the formula (I) in which L represents L-1, where R² has the especially preferred meanings given above.

Emphasis is given to compounds of the formula (I) in which L represents L-2.

Emphasis is given to compounds of the formula (I) in which R¹ represents hydrogen.

Emphasis is given to compounds of the formula (I) in which R¹ represents formyl.

15 Emphasis is furthermore given to compounds of the formula (I) in which R¹ represents -C(=O)C(=O)R⁴ where R⁴ is as defined above.

Emphasis is given to compounds of the formula (I) in which A represents A1.

Saturated or unsaturated hydrocarbon radicals, such as alkyl or alkenyl, can in each case be straight-chain or branched, as far as this is possible, including in combination with heteroatoms, such as, for 20 example, in alkoxy.

Optionally substituted radicals can be mono- or polysubstituted, where in the case of polysubstitution the substituents can be identical or different.

25 Halogen-substituted radicals, such as, for example, haloalkyl, are mono- or polyhalogenated. In the case of polyhalogenation, the halogen atoms can be identical or different. Here, halogen represents fluorine, chlorine, bromine and iodine, in particular fluorine, chlorine and bromine.

30 However, the general or preferred radical definitions or illustrations given above can also be combined with one another as desired, i.e. between the respective ranges and preferred ranges. The definitions apply both to the end products and, correspondingly, to the precursors and intermediates.

The definitions mentioned can be combined with one another as desired. Moreover, individual definitions may not apply.

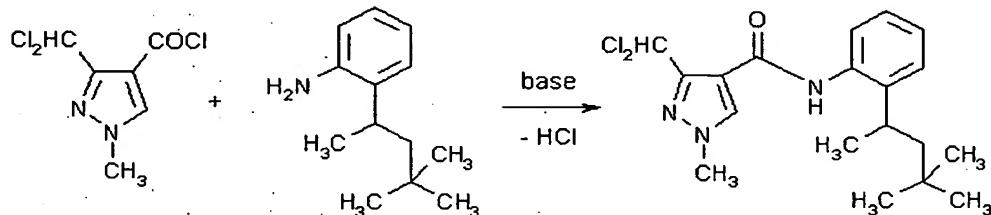
35

Preference, particular preference or very particular preference is given to the compounds of the formula (I) which carry the substituents mentioned as being preferred, particularly preferred and very particularly preferred, respectively.

Description of the processes according to the invention for preparing the hexylcarboxanilides of the formula (I) and the intermediates

Process (a)

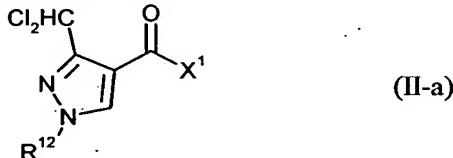
5 Using 3-dichloromethyl-1-methyl-1H-pyrazole-4-carbonyl chloride and [2-(1,3,3-trimethylbutyl)-phenyl]amine as starting materials, the process (a) according to the invention can be illustrated by the formula scheme below:



10 Formula (II) provides a general definition of the carboxylic acid derivatives required as starting materials for carrying out the process (a) according to the invention. In this formula (II), A preferably, particularly pin connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred, respectively, for A. X¹ preferably represents chlorine, bromine or hydroxyl.

15 Most of the carboxylic acid derivatives of the formula (II) are known and/or can be prepared by known processes (cf. WO 93/11117, EP-A 0 545 099, EP-A 0 589 301 and EP-A 0 589 313).

3-Dichloromethyl-1H-pyrazole-4-carboxylic acid derivatives of the formula (II-a)

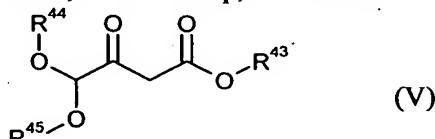


20 in which

R¹² is as defined above,

X¹ represents halogen or hydroxyl

can be obtained when, in a first step, ketoacetals of the formula (V)



25

in which

R⁴³ represents C₁-C₄-alkyl, preferably methyl, ethyl, n-, isopropyl, n-, sec-, tert-butyl,

R⁴⁴ and R⁴⁵ each represent methyl or ethyl, or

R⁴⁴ and R⁴⁵ together represent -(CH₂)₃- or -CH₂-C(CH₃)₂-CH₂-

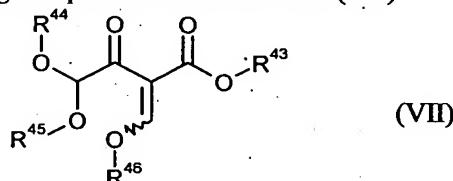
are reacted with alkyl orthoformates of the formula (VI)



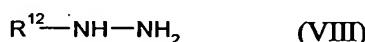
in which

5 R^{46} represents $\text{C}_1\text{-C}_4$ -alkyl, preferably methyl, ethyl, n-, isopropyl, n-, sec-, tert-butyl
in the presence of an anhydride (for example acetic anhydride)

and the resulting compounds of the formula (VII)



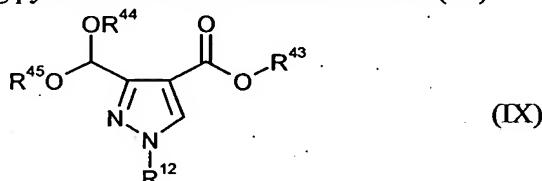
10 in which R^{43} , R^{44} , R^{45} and R^{46} are as defined above,
are in a second step, reacted with hydrazine derivatives of the formula (VIII)



in which R^{12} is as defined above

15 in the presence of a diluent (for example methanol)

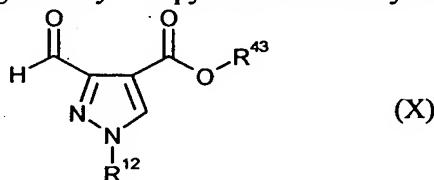
and the resulting pyrazole derivatives of the formula (IX)



in which R^{12} , R^{43} , R^{44} and R^{45} are as defined above

20 are, in a third step, reacted in the presence of an acid (for example hydrochloric acid) and in the presence of a diluent (for example dioxane)

and the resulting 3-formyl-1H-pyrazole-4-carboxylic esters of the formula (X)

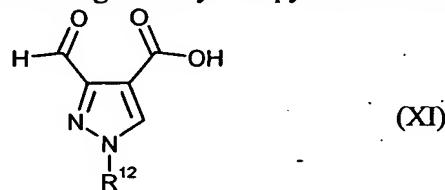


25 in which R^{12} and R^{43} are as defined above

are either

a) in a fourth step hydrolyzed in the presence of a base (for example lithium hydroxide) and in the presence of a diluent (for example tetrahydrofuran)

and the resulting 3-formyl-1H-pyrazole-4-carboxylic acids of the formula (XI)



in which R¹² is as defined above

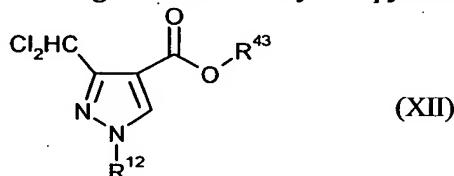
5 are then reacted with a chlorinating agent (for example phosphorus pentachloride) in the presence of a diluent (for example dichloromethane),

or

b) in a fourth step reacted with a chlorinating agent (for example phosphorus pentachloride) in the presence of a diluent (for example dichloromethane)

10

and the resulting 3-dichloromethyl-1H-pyrazole-4-carboxylic esters of the formula (XII)



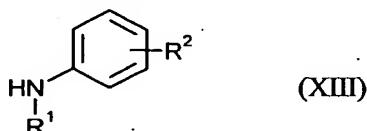
in which R¹² and R⁴³ are as defined above

15 are then hydrolyzed in the presence of a base (for example lithium hydroxide) and in the presence of a diluent (for example tetrahydrofuran).

The formula (III) provides a general definition of the aniline derivatives furthermore required as starting materials for carrying out the process (a) according to the invention. In this formula (III), L, R¹ and R³ preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred for these radicals.

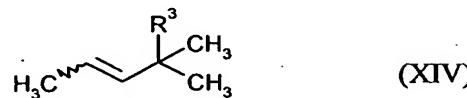
Some of the aniline derivatives of the formula (III) in which L represents L-1 are novel. Aniline 25 derivatives of formula (III) in which L represents L-1 can be prepared by

c) reacting, in a first step, an aniline derivative of the formula (XIII)



in which R¹ and R² are as defined above

30 with an alkene of the formula (XIV)

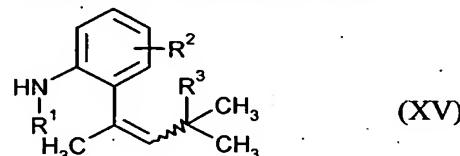


in which R³ is as defined above

in the presence of a catalyst, if appropriate in the presence of a base and if appropriate in the presence of a diluent,

5

and hydrogenating the resulting alkene aniline of the formula (XV)

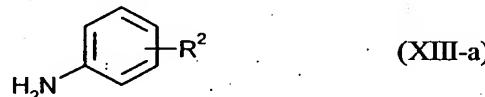


in which R¹, R² and R³ are as defined above

10 in a second step, if appropriate in the presence of a diluent and if appropriate in the presence of a catalyst.

The formula (XIII) provides a general definition of the aniline derivatives required as starting materials for carrying out the process (c) according to the invention. In this formula (XIII), R¹ and R² preferably, particularly preferably and very particularly preferably have those meanings which have 15 already been mentioned in connection with the description of the compounds of the formula (I) according to the invention as being preferred, particularly preferred and very particularly preferred for these radicals.

Aniline derivatives of the formula (XIII) are known or can be obtained by known methods. Aniline 20 derivatives of the formula (XIII) in which R¹ does not represent hydrogen can be obtained by reacting anilines of the formula (XIII-a)



in which R² is as defined above

with halides of the formula (IV)

25 $R^{1-A} - X^2$ (IV)

in which R^{1-A} and X² are as defined above

in the presence of a base and in the presence of a diluent. [The reaction conditions of process (b) apply correspondingly.]

30 The formula (XIV) provides a general definition of the alkenes furthermore required as starting materials for carrying out the process (c) according to the invention. In this formula (XIV), R³ preferably, particularly preferably and very particularly preferably has those meanings which have already been mentioned in connection with the description of the compounds of the formula (I)

according to the invention as being preferred, particularly preferred and very particularly preferred for this radical.

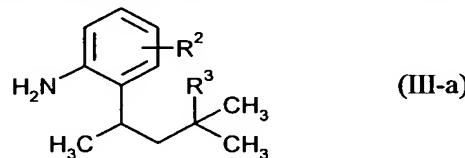
Alkenes of the formula (XIV) are known or can be obtained by known methods.

5

The formula (XV) provides a general definition of the alkene anilines which are intermediates obtained when carrying out the process (c) according to the invention. In this formula (XV), R¹, R² and R³ preferably, particularly preferably and very particularly preferably have those meanings which have already been mentioned in connection with the description of the compounds of the formula (I) 10 according to the invention as being preferred, particularly preferred and very particularly preferred for these radicals.

Some of the alkene anilines of the formula (XV) are known.

15 The process (c) according to the invention can be carried out in various variants. Thus, it is possible to initially react anilines of the formula (XIII-a) with alkenes of the formula (XIV) to give the corresponding aniline derivatives of the formula (III-a)



in which R² and R³ are as defined above,

20 which are then, if appropriate, reacted with halides of the formula (IV)

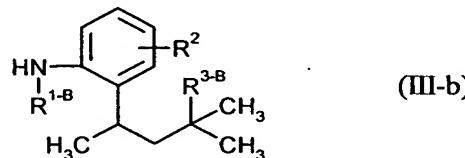


in which R^{1-A} and X² are as defined above

in the presence of a base and in the presence of a diluent to give the corresponding aniline derivatives 25 of the formula (III). [The reaction conditions of process (b) apply correspondingly.]

However, it is also possible to carry out the reaction with a halide of the formula (IV) at the stage of the alkene anilines of the formula (XV) and to hydrogenate afterwards.

30 Aniline derivatives of the formula (III-b)



in which

a) R^{1-B} represents hydrogen and

R^{3-B} represents halogen, C₃-C₈-alkyl, C₁-C₈-haloalkyl,
or
b) R^{1-B} represents C₁-C₈-alkyl, C₁-C₆-alkylsulphanyl, C₁-C₆-alkylsulphonyl, C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-cycloalkyl; C₁-C₆-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄-haloalkylsulphanyl, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₄-alkoxy-C₁-C₄-alkyl, C₃-C₈-halocycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl, formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl; halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in each case 1 to 13 fluorine, chlorine and/or bromine atoms;
(C₁-C₈-alkyl)carbonyl, (C₁-C₈-alkoxy)carbonyl, (C₁-C₄-alkoxy-C₁-C₄-alkyl)carbonyl, (C₃-C₈-cycloalkyl)carbonyl; (C₁-C₆-haloalkyl)carbonyl, (C₁-C₆-haloalkoxy)carbonyl, (halo-C₁-C₄-alkoxy-C₁-C₄-alkyl)carbonyl, (C₃-C₈-halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O)R⁴, -CONR⁵R⁶ or -CH₂NR⁷R⁸, and
R^{3-B} represents hydrogen, halogen, C₁-C₈-alkyl, C₁-C₈-haloalkyl,

and

R², R⁴, R⁵, R⁶, R⁷ and R⁸ are each as defined above

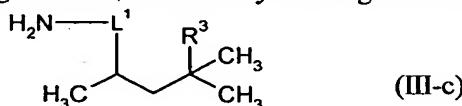
are novel and also form part of the subject-matter of this application.

The preferred, particularly preferred and very particularly preferred meanings of R¹ and R³ apply correspondingly to R^{1-B} and R^{3-B}, where in case a) R^{1-B} always represents hydrogen and R^{3-B} does not represent hydrogen, methyl or ethyl and in case b) R^{1-B} does not represent hydrogen. The preferred, particularly preferred and very particularly preferred meanings of R², R⁴, R⁵, R⁶, R⁷ and R⁸ also apply to the novel compounds of the formula (III-b).

Emphasis is given to compounds of the formula (III-b) in which R¹ and R² each represent hydrogen and R³ represents fluorine, chlorine, methyl, ethyl, trifluoromethyl or pentafluoroethyl.

The aniline derivatives of the formula (III) in which L represents L-2, L-3 or L-4 are known and/or can be obtained by known processes (cf., for example, EP-A 1 036 793 and EP-A 0 737 682).

Aniline derivatives of the formula (III) in which L represents L-2, L-3 or L-4 and R¹ does not represent hydrogen can be obtained by reacting anilines of the formula (III-c)



in which

L¹ represents L-2, L-3 or L-4 and

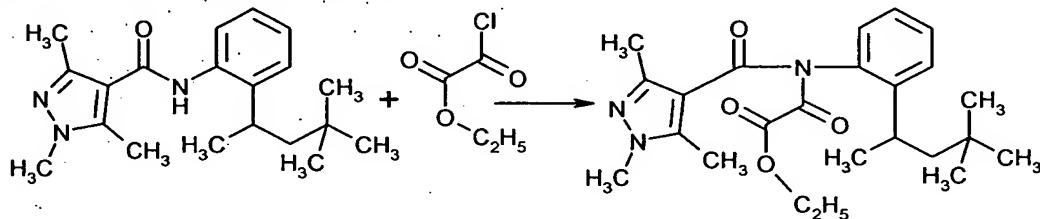
L-2, L-3, L-4 and R³ are as defined above
with halides of the formula (IV)



5 in which R^{1-A} and X² are as defined above
in the presence of a base and in the presence of a diluent. [The reaction conditions of process (b)
apply correspondingly.]

Process (b)

10 Using 1,3,5-trimethyl-N-[2-(1,3,3-trimethylbutyl)phenyl]-1H-pyrazole-4-carboxamide and ethyl
chloro(oxo)acetate as starting materials, the course of the process (b) according to the invention can
be illustrated by the formula scheme below:



15 The formula (I-a) provides a general definition of the hexylcarboxanilides required as starting materials
for carrying out the process (b) according to the invention. In this formula (I-a), R², R³ and A preferably,
particularly preferably and very particularly preferably have those meanings which have already been
mentioned in connection with the description of the compounds of the formula (I) according to the
invention as being preferred, particularly preferred and very particularly preferred, respectively, for
these radicals.

20 The hexylcarboxanilides of the formula (I-a) are also compounds according to the invention and form
part of the subject-matter of this application. They can be obtained by process (a) according to the
invention (where R¹ = hydrogen).

25 The formula (IV) provides a general definition of the halides furthermore required for carrying out
the process (b) according to the invention.

30 R^{1-A} preferably represents C₁-C₆-alkyl, C₁-C₄-alkylsulphanyl, C₁-C₄-alkylsulphonyl, C₁-C₃-alkoxy-
C₁-C₃-alkyl, C₃-C₆-cycloalkyl; C₁-C₄-haloalkyl, C₁-C₄-haloalkylthio, C₁-C₄-halo-
alkylsulphanyl, C₁-C₄-haloalkylsulphonyl, halo-C₁-C₃-alkoxy-C₁-C₃-alkyl, C₃-C₈-halo-
cycloalkyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; formyl,
formyl-C₁-C₃-alkyl, (C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, (C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl;
halo-(C₁-C₃-alkyl)carbonyl-C₁-C₃-alkyl, halo-(C₁-C₃-alkoxy)carbonyl-C₁-C₃-alkyl having in
each case 1 to 13 fluorine, chlorine and/or bromine atoms;

(C₁-C₆-alkyl)carbonyl, (C₁-C₄-alkoxy)carbonyl, (C₁-C₃-alkoxy-C₁-C₃-alkyl)carbonyl, (C₃-C₆-cycloalkyl)carbonyl; (C₁-C₄-haloalkyl)carbonyl, (C₁-C₄-haloalkoxy)carbonyl, (halo-C₁-C₃-alkoxy-C₁-C₃-alkyl)carbonyl, (C₃-C₆-halocycloalkyl)carbonyl having in each case 1 to 9 fluorine, chlorine and/or bromine atoms; or -C(=O)C(=O)R⁴, -CONR⁵R⁶ or -CH₂NR⁷R⁸.

5 R^{1-A} particularly preferably represents methyl, ethyl, n- or isopropyl, n-, iso-, sec- or tert-butyl, pentyl or hexyl, methylsulphanyl, ethylsulphanyl, n- or isopropylsulphanyl, n-, iso-, sec- or tert-butylsulphanyl, methylsulphonyl, ethylsulphonyl, n- or isopropylsulphonyl, n-, iso-, sec- or tert-butylsulphonyl, methoxymethyl, methoxyethyl, ethoxymethyl, ethoxyethyl, cyclopropyl, cyclopentyl, cyclohexyl, trifluoromethyl, trichloromethyl, trifluoroethyl, difluoromethylthio, difluorochloromethylthio, trifluoromethylthio, trifluoromethylsulphanyl, trifluoromethylsulphonyl, trifluoromethoxymethyl; formyl, -CH₂-CHO, -(CH₂)₂-CHO, -CH₂-CO-CH₃, -CH₂-CO-CH₂CH₃, -CH₂-CO-CH(CH₃)₂, -(CH₂)₂-CO-CH₃, -(CH₂)₂-CO-CH₂CH₃, -(CH₂)₂-CO-CH(CH₃)₂, -(CH₂)₂-CO₂CH₃, -CH₂-CO₂CH₂CH₃, -CH₂-CO₂CH(CH₃)₂, -CH₂-CO-CF₃, -CH₂-CO-CCl₃, -CH₂-CO-CH₂CF₃, -CH₂-CO-CH₂CCl₃, -(CH₂)₂-CO-CH₂CF₃, -(CH₂)₂-CO-CH₂CCl₃, -CH₂-CO₂CCl₂CCl₃, -(CH₂)₂-CO₂CH₂CF₃, -(CH₂)₂-CO₂CF₂CF₃, -(CH₂)₂-CO₂CH₂CCl₃, -(CH₂)₂-CO₂CCl₂CCl₃; methylcarbonyl, ethylcarbonyl, n-propylcarbonyl, isopropylcarbonyl, tert-butylcarbonyl, methoxycarbonyl, ethoxycarbonyl, tert-butoxycarbonyl, cyclopropylcarbonyl; trifluoromethylcarbonyl, trifluoromethoxycarbonyl, or -C(=O)C(=O)R⁵, -CONR⁶R⁷ or -CH₂NR⁸R⁹.

10 R^{1-A} very particularly preferably represents methyl, methoxymethyl, formyl, -CH₂-CHO, -(CH₂)₂-CHO, -CH₂-CO-CH₃, -CH₂-CO-CH₂CH₃, -CH₂-CO-CH(CH₃)₂, -C(=O)CHO, -C(=O)C(=O)CH₃, -C(=O)C(=O)CH₂OCH₃, -C(=O)CO₂CH₃, -C(=O)CO₂CH₂CH₃.

15 X² preferably represents chlorine or bromine.

Halides of the formula (IV) are known.

20

Reaction conditions

Suitable diluents for carrying out the process (a) according to the invention are all inert organic solvents. These preferably include aliphatic, alicyclic or aromatic hydrocarbons, such as, for example, petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane, benzene, toluene, xylene or decalin; halogenated hydrocarbons, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; ethers, such as diethyl ether, diisopropyl ether, methyl tert-butyl ether, methyl tert-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole, or amides, such as N,N-

25

dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide.

5 The process (a) according to the invention is, if appropriate, carried out in the presence of a suitable acid acceptor. Suitable acid acceptors are all customary inorganic or organic bases. These preferably include include alkaline earth metal or alkali metal hydrides, hydroxides, amides, alkoxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydride, sodium amide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, 10 potassium carbonate, potassium bicarbonate, sodium bicarbonate or ammonium carbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, N,N-dimethylbenzylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

15 The process (a) according to the invention is, if appropriate, carried out in the presence of a suitable condensing agent. Suitable condensing agents are all condensing agents customarily used for such amidation reactions. Acid halide formers, such as phosgene, phosphorus tribromide, phosphorus trichloride, phosphorus pentachloride, phosphorus oxychloride or thionyl chloride; anhydride formers, such as ethyl chloroformate, methyl chloroformate, isopropyl chloroformate, isobutyl chloroformate or methanesulphonyl chloride; carbodiimides, such as N,N'-dicyclohexylcarbodiimide (DCC), or other customary condensing agents, such as phosphorus pentoxide, polyphosphoric acid, N,N'-carbonyldiimidazole, 2-ethoxy-N-ethoxycarbonyl-1,2-dihydroquinoline (EEDQ), triphenylphosphine/carbon tetrachloride or bromotripyrrolidinophosphonium hexafluorophosphate 20 may be mentioned by way of example.

25

The process (a) is, if appropriate, carried out in the presence of a catalyst. Examples which may be mentioned are 4-dimethylaminopyridine, 1-hydroxybenzotriazole or dimethylformamide.

30 When carrying out the process (a) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the process is carried out at temperatures of from 0°C to 150°C, preferably at temperatures of from 0°C to 80°C.

35 For carrying out process (a) according to the invention for preparing the compounds of the formula (I), in general from 0.2 to 5 mol, preferably from 0.5 to 2 mol, of aniline derivative of the formula (III) are employed per mole of the carboxylic acid derivative of the formula (II).

Suitable diluents for carrying out the process (b) according to the invention are all inert organic solvents. These preferably include aliphatic, alicyclic or aromatic hydrocarbons, such as, for example,

petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane, benzene, toluene, xylene or decalin; halogenated hydrocarbons, such as, for example, chlorobenzene, dichlorobenzene, dichloromethane, chloroform, carbon tetrachloride, dichloroethane or trichloroethane; ethers, such as diethyl ether, diisopropyl ether, methyl tert-butyl ether, methyl tert-amyl ether, dioxane, 5 tetrahydrofuran, 1,2-dimethoxyethane, 1,2-diethoxyethane or anisole, or amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide.

10 The process (b) according to the invention is carried out in the presence of a base. Suitable bases are all customary inorganic or organic bases. These preferably include alkaline earth metal or alkali metal hydrides, hydroxides, amides, alkoxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydride, sodium amide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate, sodium 15 bicarbonate or caesium carbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, N,N-dimethylbenzylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

20 When carrying out the process (b) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the process is carried out at temperatures of from 0°C to 150°C, preferably at temperatures of from 20°C to 110°C.

25 For carrying out the process (b) according to the invention for preparing the compounds of the formula (I), in general from 0.2 to 5 mol, preferably from 0.5 to 2 mol, of halide of the formula (IV) are employed per mole of the hexylcarboxanilide of the formula (I-a).

30 Suitable diluents for carrying out the first step of the process (c) according to the invention are all inert organic solvents. These preferably include nitriles, such as acetonitrile, propionitrile, n- or i-butyronitrile or benzonitrile, or amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide, N-methylpyrrolidone or hexamethylphosphoric triamide.

35 The first step of the process (c) according to the invention is, if appropriate, carried out in the presence of a suitable acid acceptor. Suitable acid acceptors are all customary inorganic or organic bases. These preferably include alkaline earth metal or alkali metal hydrides, hydroxides, amides, alkoxides, acetates, carbonates or bicarbonates, such as, for example, sodium hydride, sodium amide, sodium methoxide, sodium ethoxide, potassium tert-butoxide, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium bicarbonate, sodium bicarbonate or

ammonium carbonate, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, N,N-dimethylaniline, N,N-dimethylbenzylamine, pyridine, N-methylpiperidine, N-methylmorpholine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

5

The first step of the process (c) according to the invention is carried out in the presence of one or more catalysts.

These are in particular palladium salts or complexes. Preferred for this purpose are palladium chloride, palladium acetate, tetrakis(triphenylphosphine)palladium or bis(triphenylphosphine)-

10 palladium dichloride. It is also possible to generate a palladium complex in the reaction mixture by adding a palladium salt and a complex ligand separately to the reaction.

Preferred ligands are organophosphorus compounds. Examples which may be mentioned are:

15 triphenylphosphine, tri-o-tolylphosphine, 2,2'-bis(diphenylphosphino)-1,1'-binaphthyl, dicyclohexylphosphinebiphenyl, 1,4-bis(diphenylphosphino)butane, bisdiphenylphosphinoferrocene, di-(tert-butylphosphino)biphenyl, di(cyclohexylphosphino)biphenyl, 2-dicyclohexylphosphino-2'-N,N-dimethylaminobiphenyl, tricyclohexylphosphine, tri-tert-butylphosphine. However, it is also possible to dispense with ligands.

20 The first step of the process (c) according to the invention is furthermore, if appropriate, carried out in the presence of a further metal salt, such as copper salts, for example copper(I) iodide.

When carrying out the first step of the process (c) according to the invention, the reaction

25 temperatures can be varied within a relatively wide range. In general, the reaction is carried out at temperatures of from 20°C to 180°C, preferably at temperatures of from 50°C to 150°C.

For carrying out the first step of the process (c) according to the invention for preparing the alkene anilines of the formula (XV), in general from 1 to 5 mol, preferably from 1 to 3 mol, of alkene of the formula (XIV) are employed per mole of the aniline derivative of the formula (XIII).

30

Suitable diluents for carrying out the second step (hydrogenation) of the process (c) according to the invention are all inert organic solvents. These preferably include aliphatic or alicyclic hydrocarbons, such as, for example, petroleum ether, hexane, heptane, cyclohexane, methylcyclohexane or decalin;

35 ethers, such as diethyl ether, diisopropyl ether, methyl tert-butyl ether, methyl tert-amyl ether, dioxane, tetrahydrofuran, 1,2-dimethoxyethane or 1,2-diethoxyethane; alcohols, such as methanol, ethanol, n- or isopropanol, n-, iso-, sec- or tert-butanol, ethanediol, propane-1,2-diol, ethoxyethanol,

methoxyethanol, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, mixtures thereof with water or pure water.

The second step (hydrogenation) of the process (c) according to the invention is carried out in the presence of a catalyst. Suitable catalysts are all catalysts which are customarily used for hydrogenations. Examples which may be mentioned are: Raney nickel, palladium or platinum, if appropriate on a support, such as, for example activated carbon.

5 Instead of in the presence of hydrogen in combination with a catalyst, the hydrogenation in the second step of the process (c) according to the invention can also be carried out in the presence of triethylsilane.

10 When carrying out the second step of the process (c) according to the invention, the reaction temperatures can be varied within a relatively wide range. In general, the reaction is carried out at temperatures of from 0°C to 150°C, preferably at temperatures of from 20°C to 100°C.

15 The second step of the process (c) according to the invention is carried out under a hydrogen pressure between 0.5 and 200 bar, preferably between 2 and 50 bar, particularly preferably between 3 and 10 bar.

20 Unless indicated otherwise, all processes according to the invention are generally carried out under atmospheric pressure. However, it is also possible to operate under elevated or reduced pressure – in general between 0.1 bar and 10 bar.

25 The substances according to the invention have potent microbial activity and can be employed for controlling unwanted microorganisms, such as fungi and bacteria, in crop protection and in the protection of materials.

30 Fungicides can be employed in crop protection for controlling Plasmodiophoromycetes, Oomycetes, Chytridiomycetes, Zygomycetes, Ascomycetes, Basidiomycetes and Deuteromycetes.

Bactericides can be employed in crop protection for controlling Pseudomonadaceae, Rhizobiaceae, Enterobacteriaceae, Corynebacteriaceae and Streptomycetaceae.

35 Some pathogens causing fungal and bacterial diseases which come under the generic names listed above may be mentioned as examples, but not by way of limitation:

Xanthomonas species, such as, for example, Xanthomonas campestris pv. oryzae;

Pseudomonas species, such as, for example, Pseudomonas syringae pv. lachrymans;

Erwinia species, such as, for example, Erwinia amylovora;

Pythium species, such as, for example, Pythium ultimum;

Phytophthora species, such as, for example, Phytophthora infestans;

Pseudoperonospora species, such as, for example, Pseudoperonospora humuli or

Pseudoperonospora cubensis;

Plasmopara species, such as, for example, *Plasmopara viticola*;
Bremia species, such as, for example, *Bremia lactucae*;
Peronospora species, such as, for example, *Peronospora pisi* or *P. brassicae*;
Erysiphe species, such as, for example, *Erysiphe graminis*;
5 Sphaerotheca species, such as, for example, *Sphaerotheca fuliginea*;
Podosphaera species, such as, for example, *Podosphaera leucotricha*;
Venturia species, such as, for example, *Venturia inaequalis*;
Pyrenophora species, such as, for example, *Pyrenophora teres* or *P. graminea*
(conidia form: *Drechslera*, syn: *Helminthosporium*);
10 Cochliobolus species, such as, for example, *Cochliobolus sativus*
(conidia form: *Drechslera*, syn: *Helminthosporium*);
Uromyces species, such as, for example, *Uromyces appendiculatus*;
Puccinia species, such as, for example, *Puccinia recondita*;
Sclerotinia species, such as, for example, *Sclerotinia sclerotiorum*;
15 Tilletia species, such as, for example, *Tilletia caries*;
Ustilago species, such as, for example, *Ustilago nuda* or *Ustilago avenae*;
Pellicularia species, such as, for example, *Pellicularia sasakii*;
Pyricularia species, such as, for example, *Pyricularia oryzae*;
Fusarium species, such as, for example, *Fusarium culmorum*;
20 Botrytis species, such as, for example, *Botrytis cinerea*;
Septoria species, such as, for example, *Septoria nodorum*;
Leptosphaeria species, such as, for example, *Leptosphaeria nodorum*;
Cercospora species, such as, for example, *Cercospora canescens*;
Alternaria species, such as, for example, *Alternaria brassicae*; and
25 Pseudocercospora species, such as, for example, *Pseudocercospora herpotrichoides*,
Rhizoctonia species, such as, for example, *Rhizoctonia solani*.

The active compounds according to the invention also show a strong invigorating action in plants.
Accordingly, they are suitable for mobilizing the internal defences of the plant against attack by
30 unwanted microorganisms.

In the present context, plant-invigorating (resistance-inducing) compounds are to be understood as meaning substances which are capable of stimulating the defence system of plants such that, when the treated plants are subsequently inoculated with unwanted microorganisms, they display substantial resistance to these microorganisms.
35

In the present case, unwanted microorganisms are to be understood as meaning phytopathogenic fungi, bacteria and viruses. The compounds according to the invention can thus be used to protect plants within a certain period of time after treatment against attack by the pathogens mentioned.

The period of time for which this protection is achieved generally extends for 1 to 10 days, preferably 1 to 7 days, from the treatment of the plants with the active compounds.

5 The fact that the active compounds are well tolerated by plants at the concentrations required for controlling plant diseases permits the treatment of above-ground parts of plants, of propagation stock and seeds, and of the soil.

10 Here, the active compounds according to the invention can be used with particularly good results for controlling cereal diseases, such as, for example, against *Puccinia* species, and of diseases in viticulture and in the cultivation of fruits and vegetables, such as, for example, against *botrytis*, *Venturia* or *Alternaria* species.

15 The active compounds according to the invention are also suitable for increasing the yield of crops. In addition, they show reduced toxicity and are well tolerated by plants.

20 If appropriate, the active compounds according to the invention can, at certain concentrations and application rates, also be employed as herbicides, for regulating plant growth and for controlling animal pests. If appropriate, they can also be used as intermediates or precursors in the synthesis of other active compounds.

25 According to the invention, it is possible to treat all plants and parts of plants. Plants are to be understood here as meaning all plants and plant populations, such as desired and undesired wild plants or crop plants (including naturally occurring crop plants). Crop plants can be plants which can be obtained by conventional breeding and optimization methods or by biotechnological and genetic engineering methods or combinations of these methods, including the transgenic plants and including plant cultivars which can or cannot be protected by plant breeders' certificates. Parts of plants are to be understood as meaning all above-ground and below-ground parts and organs of plants, such as shoot, leaf, flower and root, examples which may be mentioned being leaves, needles, stems, trunks, flowers, fruit-bodies, fruits and seeds and also roots, tubers and rhizomes.

30 Parts of plants also include harvested material and vegetative and generative propagation material, for example seedlings, tubers, rhizomes, cuttings and seeds.

35 The treatment of the plants and parts of plants according to the invention with the active compounds is carried out directly or by action on their environment, habitat or storage area according to customary treatment methods, for example by dipping, spraying, evaporating, atomizing, broadcasting, brushing-on and, in the case of propagation material, in particular in the case of seeds, furthermore by one- or multilayer coating.

In the protection of materials, the compounds according to the invention can be employed for protecting industrial materials against infection with, and destruction by, unwanted microorganisms.

5 Industrial materials in the present context are understood as meaning non-living materials which have been prepared for use in industry. For example, industrial materials which are intended to be protected by active compounds according to the invention from microbial change or destruction can be tackifiers, sizes, paper and board, textiles, leather, wood, paints and plastic articles, cooling lubricants and other materials which can be infected with, or destroyed by, microorganisms. Parts
10 of production plants, for example cooling-water circuits, which may be impaired by the proliferation of microorganisms may also be mentioned within the scope of the materials to be protected. Industrial materials which may be mentioned within the scope of the present invention are preferably tackifiers, sizes, paper and board, leather, wood, paints, cooling lubricants and heat-transfer liquids, particularly preferably wood.

15

Microorganisms capable of degrading or changing the industrial materials which may be mentioned are, for example, bacteria, fungi, yeasts, algae and slime organisms. The active compounds according to the invention preferably act against fungi, in particular moulds, wood-discolouring and wood-destroying fungi (Basidiomycetes) and against slime organisms and algae.

20

Microorganisms of the following genera may be mentioned as examples:

Alternaria, such as *Alternaria tenuis*,

Aspergillus, such as *Aspergillus niger*,

Chaetomium, such as *Chaetomium globosum*,

25 Coniophora, such as *Coniophora puteana*,

Lentinus, such as *Lentinus tigrinus*,

Penicillium, such as *Penicillium glaucum*,

Polyporus, such as *Polyporus versicolor*,

Aureobasidium, such as *Aureobasidium pullulans*,

30 Sclerophoma, such as *Sclerophoma pityophila*,

Trichoderma, such as *Trichoderma viride*,

Escherichia, such as *Escherichia coli*,

Pseudomonas, such as *Pseudomonas aeruginosa*, and

Staphylococcus, such as *Staphylococcus aureus*.

35

Depending on their particular physical and/or chemical properties, the active compounds can be converted into the customary formulations, such as solutions, emulsions, suspensions, powders, foams, pastes, granules, aerosols and microencapsulations in polymeric substances and in coating compositions for seeds, and ULV cool and warm fogging formulations.

These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is liquid solvents, liquefied gases under pressure, and/or solid carriers, optionally with the use of surfactants, that is emulsifiers and/or dispersants, and/or foam formers.

5 If the extender used is water, it is also possible to employ, for example, organic solvents as auxiliary solvents. Essentially, suitable liquid solvents are: aromatics such as xylene, toluene or alkylnaphthalenes, chlorinated aromatics or chlorinated aliphatic hydrocarbons such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons such as cyclohexane or paraffins, for example petroleum fractions, alcohols such as butanol or glycol and their ethers and esters, ketones such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents such as dimethylformamide or dimethyl sulphoxide, or else water. Liquefied gaseous extenders or carriers are to be understood as meaning liquids which are gaseous at standard temperature and under atmospheric pressure, for example aerosol propellants such as halogenated hydrocarbons, or else butane, propane, nitrogen and carbon dioxide. Suitable solid carriers are: for example ground natural minerals such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals such as finely divided silica, alumina and silicates. Suitable solid carriers for granules are: for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, or else synthetic granules of inorganic and organic meals, and granules of organic material such as 10 sawdust, coconut shells, maize cobs and tobacco stalks. Suitable emulsifiers and/or foam formers are: for example nonionic and anionic emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates, or else protein hydrolysates. Suitable dispersants are: for example lignosulphite waste liquors and methylcellulose.

15

20

25 Tackifiers such as carboxymethylcellulose, natural and synthetic polymers in the form of powders, granules or latices, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, or else natural phospholipids such as cephalins and lecithins and synthetic phospholipids can be used in the formulations. Other possible additives are mineral and vegetable oils.

30 It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

35 The formulations generally comprise between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

The active compounds according to the invention can, as such or in their formulations, also be used in a mixture with known fungicides, bactericides, acaricides, nematicides or insecticides, to broaden, for example, the activity spectrum or to prevent development of resistance. In many cases, synergistic effects are obtained, i.e. the activity of the mixture is greater than the activity of the individual components.

Suitable mixing components are, for example, the following compounds:

Fungicides:

2-phenylphenol; 8-hydroxyquinoline sulphate; acibenzolar-S-methyl; aldimorph; amidoflumet; 10 ampropylfos; ampropylfos-potassium; andoprim; anilazine; azaconazole; azoxystrobin; benalaxyl; benodanil; benomyl; benthiavalicarb-isopropyl; benzamacril; benzamacril-isobutyl; bilanafos; binapacryl; biphenyl; bitertanol; blasticidin-S; bromuconazole; bupirimate; buthiobate; butylamine; calcium polysulphide; capsimycin; captafol; captan; carbendazim; carboxin; carpropamid; carvone; chinomethionat; chlobenthiazole; chlorsenazole; chloroneb; chlorothalonil; 15 chlozolinate; clozylacon; cyazofamid; cyflufenamid; cymoxanil; cyproconazole; cyprodinil; cyprofuram; Dagger G; debacarb; dichlofluanid; dichlone; dichlorophen; diclocymet; diclomezine; dicloran; diethofencarb; difenoconazole; diflumetorim; dimethirimol; dimethomorph; dimoxystrobin; diniconazole; diniconazole-M; dinocap; diphenylamine; dipyridithione; ditalimfos; dithianon; dodine; drazoxolon; edifenphos; epoxiconazole; ethaboxam; ethirimol; etridiazole; 20 famoxadone; fenamidone; fenapanil; fenarimol; fenbuconazole; fenfuram; fenhexamid; fenitropan; fenoxanil; fenpiclonil; fenpropidin; fenpropimorph; ferbam; fluazinam; flubenzimine; fludioxonil; flumetover; flumorph; fluoromide; fluoxastrobin; fluquinconazole; flurprimidol; flusilazole; flusulphamide; flutolanil; flutriafol; folpet; fosetyl-Al; fosetyl-sodium; fuberidazole; furalaxyl; furametpyr; furcarbanil; furmecyclox; guazatine; hexachlorobenzene; hexaconazole; hymexazole; 25 imazalil; imibenconazole; iminoctadine triacetate; iminoctadine tris(albesil); iodocarb; ipconazole; iprobenfos; iprodione; iprovalicarb; irumamycin; isoprothiolane; isovaledione; kasugamycin; kresoxim-methyl; mancozeb; maneb; meferimzone; mepanipyrim; mepronil; metalaxyl; métalaxyl-M; metconazole; methasulphocarb; methfuroxam; metiram; metominostrobin; metsulphovax; mildiomycin; myclobutanil; myclozolin; natamycin; nicobifen; nitrothal-isopropyl; noviflumuron; 30 nuarimol; ofurace; orysastrobin; oxadixyl; oxolinic acid; oxpoconazole; oxycarboxin; oxyfenthiin; paclbutrazole; pefurazoate; penconazole; pencycuron; phosdiphen; phthalide; picoxystrobin; piperalin; polyoxins; polyoxorim; probenazole; prochloraz; procymidone; propamocarb; propanosine-sodium; propiconazole; propineb; proquinazid; prothioconazole; pyraclostrobin; pyrazophos; pyrifenoxy; pyrimethanil; pyroquilon; pyroxyfur; pyrrolenitrine; quinconazole; 35 quinoxyfen; quintozene; simeconazole; spiroxamine; sulphur; tebuconazole; tecloftalam; tecnazene; tetcyclacis; tetraconazole; thiabendazole; thicyofen; thifluzamide; thiophanate-methyl; thiram; tioxymid; tolclofos-methyl; tolylfluanid; triadimefon; triadimenol; triazbutil; triazoxide; tricyclamide; tricyclazole; tridemorph; trifloxystrobin; triflumizole; triforine; triticonazole; uniconazole; validamycin A; vinclozolin; zineb; ziram; zoxamide; (2S)-N-[2-[4-[[3-(4-

chlorophenyl)-2-propynyl]oxy]-3-methoxyphenyl]ethyl]-3-methyl-2-[(methylsulphonyl)amino]-butanamide; 1-(1-naphthalenyl)-1H-pyrrole-2,5-dione; 2,3,5,6-tetrachloro-4-(methylsulphonyl)-pyridine; 2-amino-4-methyl-N-phenyl-5-thiazolecarboxamide; 2-chloro-N-(2,3-dihydro-1,1,3-trimethyl-1H-inden-4-yl)-3-pyridinecarboxamide; 3,4,5-trichloro-2,6-pyridinedicarbonitrile; 5 actinovate; cis-1-(4-chlorophenyl)-2-(1H-1,2,4-triazol-1-yl)cycloheptanol; methyl 1-(2,3-dihydro-2,2-dimethyl-1H-inden-1-yl)-1H-imidazole-5-carboxylate; monopotassium carbonate; N-(6-methoxy-3-pyridinyl)-cyclopropanecarboxamide; N-butyl-8-(1,1-dimethylethyl)-1-oxaspiro-[4.5]decane-3-amine; sodium tetrathiocarbonate; and copper salts and preparations, such as Bordeaux mixture; copper hydroxide; copper naphthenate; copper oxychloride; copper sulphate; 10 cufraneb; copper oxide; mancopper; oxine-copper.

Bactericides:

bronopol, dichlorophen, nitrapyrin, nickel dimethyldithiocarbamate, kasugamycin, othilinone, furancarboxylic acid, oxytetracyclin, probenazole, streptomycin, tecloftalam, copper sulphate and 15 other copper preparations.

Insecticides / acaricides / nematicides:

abamectin, ABG-9008, acephate, acequinocyl, acetamiprid, acetoprole, acrinathrin, AKD-1022, AKD-3059, AKD-3088, alanycarb, aldicarb, aldoxycarb, allethrin, allethrin 1R-isomers, alpha-20 cypermethrin (alphamethrin), amidoflumet, aminocarb, amitraz, avermectin, AZ-60541, azadirachtin, azamethiphos, azinphos-methyl, azinphos-ethyl, azocyclotin, Bacillus popilliae, Bacillus sphaericus, Bacillus subtilis, Bacillus thuringiensis, Bacillus thuringiensis strain EG-2348, Bacillus thuringiensis strain GC-91, Bacillus thuringiensis strain NCTC-11821, baculoviruses, Beauveria bassiana, Beauveria tenella, benclothiaz, bendiocarb, benfuracarb, 25 bensultap, benzoximate, beta-cyfluthrin, beta-cypermethrin, bifenazate, bifenthrin, binapacryl, bioallethrin, bioallethrin-S-cyclopentyl-isomer, bioethanomethrin, biopermethrin, bioresmethrin, bistrifluron, BPMC, brofenprox, bromophos-ethyl, bromopropylate, bromfenvinfos (-methyl), BTG-504, BTG-505, bufencarb, buprofezin, butathiofos, butocarboxim, butoxycarboxim, butylpyridaben, cadusafos, camphechlor, carbaryl, carbofuran, carbophenothion, carbosulphan, 30 cartap, CGA-50439, chinomethionat, chlordane, chlordimeform, chloethocarb, chlorethoxyfos, chlorfenapyr, chlorfenvinphos, chlorfluazuron, chlormephos, chlorobenzilate, chloropicrin, chlorproxyfen, chlorpyrifos-methyl, chlorpyrifos (-ethyl), chlovaporthrin, chromafenozide, cis-cypermethrin, cis-resmethrin, cis-permethrin, clopythrin, cloethocarb, clofentezine, clothianidin, clothiazaben, codlemone, coumaphos, cyanofenphos, cyanophos, cycloprene, cycloprothrin, Cydia 35 pomonella, cyfluthrin, cyhalothrin, cyhexatin, cypermethrin, cyphenothrin (1R-trans-isomer), cyromazine, DDT, deltamethrin, demeton-S-methyl, demeton-S-methylsulphone, diafenthiuron, dialifos, diazinon, dichlofenthion, dichlorvos, dicofol, dicrotophos, dicyclanil, diflubenzuron, dimefluthrin, dimethoate, dimethylvinphos, dinobuton, dinocap, dinotefuran, diofenolan, disulphoton, docusat-sodium, dofenapyn, DOWCO-439, eflusilanate, emamectin, emamectin-

benzoate, empenthrin (1R-isomer), endosulphan, Entomophthora spp., EPN, esfenvalerate, ethiofencarb, ethiprole, ethion, ethoprophos, etofenprox, etoxazole, etrimfos, famphur, fenamiphos, fenazaquin, fenbutatin oxide, fenfluthrin, fenitrothion, fenobucarb, fenothiocarb, fenoxacrim, fenoxy carb, fenpropothrin, fenpyrad, fenpyrithrin, fenpyroximate, fensulphothion, 5 fenthion, fentrifanil, fenvalerate, fipronil, flonicamid, fluacrypyrim, fluazuron, flubenzimine, flubrocythrinate, flucycloxuron, flucythrinate, flufenprox, flumethrin, flupyrazofos, flutenzin (flufenzine), fluvalinate, fonofos, formetanate, formothion, fosmethilan, fosthiazate, fubfenprox (fluproxyfen), furathiocarb, gamma-cyhalothrin, gamma-HCH, gossyplure, grandlure, granulosis viruses, halfenprox, halofenozide, HCH, HCN-801, heptenophos, 10 hexaflumuron, hexythiazox, hydramethylnone, hydroprene, IKA-2002, imidacloprid, imiprothrin, indoxacarb, iodofenphos, iprobenfos, isazofos, isofenphos, isoprocarb, isoxathion, ivermectin, japonilure, kadethrin, nuclear polyhedrosis viruses, kinoprene, lambda-cyhalothrin, lindane, lufenuron, malathion, mecarbam, mesulphenfos, metaldehyde, metam-sodium, methacrifos, methamidophos, Metharhizium anisopliae, Metharhizium flavoviride, methidathion, methiocarb, 15 methomyl, methoprene, methoxychlor, methoxyfenozide, metofluthrin, metolcarb, metoxadiazone, mevinphos, milbemectin, milbemycin, MKI-245, MON-45700, monocrotophos, moxidectin, MTI-800, naled, NC-104, NC-170, NC-184, NC-194, NC-196, niclosamide, nicotine, nitenpyram, nithiazine, NNI-0001, NNI-0101, NNI-0250, NNI-9768, novaluron, noviflumuron, OK-5101, OK-5201, OK-9601, OK-9602, OK-9701, OK-9802, omethoate, oxamyl, oxydemeton-methyl, 20 Paecilomyces fumosoroseus, parathion-methyl, parathion (-ethyl), permethrin (cis-, trans-), petroleum, PH-6045, phenothrin (1R-trans isomer), phenthroate, phorate, phosalone, phosmet, phosphamidon, phosphocarb, phoxim, piperonyl butoxide, pirimicarb, pirimiphos-methyl, pirimiphos-ethyl, potassium oleate, prallethrin, profenofos, profluthrin, promecarb, propaphos, propargite, propetamphos, propoxur, prothios, prothoate, protrifentbute, pymetrozine, pyraclofos, 25 pyresmethrin, pyrethrum, pyridaben, pyridalyl, pyridaphenthion, pyridathion, pyrimidifen, pyriproxyfen, quinalphos, resmethrin, RH-5849, ribavirin, RU-12457, RU-15525, S-421, S-1833, salithion, sebufos, SI-0009, silafluofen, spinosad, spirodiclofen, spiomesifen, sulfluramid, sulphotep, sulprofos, SZI-121, tau-fluvalinate, tebufenozide, tebufenpyrad, tebupirimfos, teflubenzuron, tefluthrin, temephos, temivinphos, terbam, terbufos, tetrachlorvinphos, tetradifon, 30 tetramethrin, tetramethrin (1R-isomer), tetrasul, theta-cypermethrin, thiacycloprid, thiamethoxam, thiapronil, thiatriphos, thiocyclam hydrogenoxalate, thiodicarb, thiofanox, thiometon, thiosultap-sodium, thuringiensin, tolfenpyrad, tralocythrin, tralomethrin, transfluthrin, triarathene, triazamate, triazophos, triazuron, trichlophenidone, trichlorfon, Trichoderma atroviride, triflumuron, trimethacarb, vamidothion, vaniliprole, verbutin, Verticillium lecanii, WL-108477, WL-40027, YI-35 YI-5201, YI-5301, YI-5302, XMC, xylylcarb, ZA-3274, zeta-cypermethrin, zolaprofos, ZXI-8901, the compound 3-methylphenyl propylcarbamate (tsumacide Z), the compound 3-(5-chloro-3-pyridinyl)-8-(2,2,2-trifluoroethyl)-8-azabicyclo[3.2.1]octane-3-carbonitrile (CAS-Reg. No. 185982-80-3) and the corresponding 3-endo-isomer (CAS-Reg. No. 185984-60-5) (cf. WO-

96/37494, WO-98/25923), and preparations which comprise insecticidally active plant extracts, nematodes, fungi or viruses.

5 A mixture with other known active compounds, such as herbicides, or with fertilizers and growth regulators, safeners and/or semiochemicals is also possible.

In addition, the compounds of the formula (I) according to the invention also have very good antimycotic activity. They have a very broad antimycotic activity spectrum in particular against dermatophytes and yeasts, moulds and diphasic fungi (for example against *Candida* species such as 10 *Candida albicans*, *Candida glabrata*) and *Epidermophyton floccosum*, *Aspergillus* species such as *Aspergillus niger* and *Aspergillus fumigatus*, *Trichophyton* species such as *Trichophyton mentagrophytes*, *Microsporon* species such as *Microsporon canis* and *audouinii*. The list of these fungi does by no means limit the mycotic spectrum which can be covered, but is only for illustration.

15 The active compounds can be used as such, in the form of their formulations or the use forms prepared therefrom, such as ready-to-use solutions, suspensions, wettable powders, pastes, soluble powders, dusts and granules. Application is carried out in a customary manner, for example by watering, spraying, atomizing, broadcasting, dusting, foaming, spreading, etc. It is furthermore 20 possible to apply the active compounds by the ultra-low volume method, or to inject the active compound preparation or the active compound itself into the soil. It is also possible to treat the seeds of the plants.

25 When using the active compounds according to the invention as fungicides, the application rates can be varied within a relatively wide range, depending on the kind of application. For the treatment of parts of plants, the active compound application rates are generally between 0.1 and 10 000 g/ha, preferably between 10 and 1000 g/ha. For seed dressing, the active compound application rates are generally between 0.001 and 50 g per kilogram of seed, preferably between 0.01 and 10 g per kilogram of seed. For the treatment of the soil, the active compound application 30 rates are generally between 0.1 and 10 000 g/ha, preferably between 1 and 5000 g/ha.

35 As already mentioned above, it is possible to treat all plants and their parts according to the invention. In a preferred embodiment, wild plant species and plant cultivars, or those obtained by conventional biological breeding, such as crossing or protoplast fusion, and parts thereof, are treated. In a further preferred embodiment, transgenic plants and plant cultivars obtained by genetic engineering, if appropriate in combination with conventional methods (Genetically Modified Organisms), and parts thereof, are treated. The term "parts" or "parts of plants" or "plant parts" has been explained above.

Particularly preferably, plants of the plant cultivars which are in each case commercially available or in use are treated according to the invention. Plant cultivars are to be understood as meaning plants having new properties ("traits") and which have been obtained by conventional breeding, by mutagenesis or by recombinant DNA techniques. They can be cultivars, varieties, bio- or 5 genotypes.

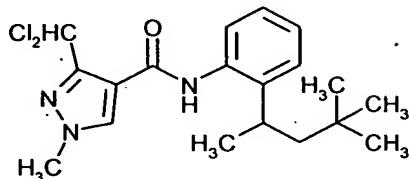
Depending on the plant species or plant cultivars, their location and growth conditions (soils, climate, vegetation period, diet), the treatment according to the invention may also result in superadditive ("synergistic") effects. Thus, for example, reduced application rates and/or a 10 widening of the activity spectrum and/or an increase in the activity of the substances and compositions which can be used according to the invention, better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest 15 yields, better quality and/or a higher nutritional value of the harvested products, better storage stability and/or processability of the harvested products are possible which exceed the effects which were actually to be expected.

The transgenic plants or plant cultivars (i.e. those obtained by genetic engineering) which are preferably to be treated according to the invention include all plants which, in the genetic 20 modification, received genetic material which imparted particularly advantageous useful properties ("traits") to these plants. Examples of such properties are better plant growth, increased tolerance to high or low temperatures, increased tolerance to drought or to water or soil salt content, increased flowering performance, easier harvesting, accelerated maturation, higher harvest yields, better quality and/or a higher nutritional value of the harvested products, better storage stability 25 and/or processability of the harvested products. Further and particularly emphasized examples of such properties are a better defence of the plants against animal and microbial pests, such as against insects, mites, phytopathogenic fungi, bacteria and/or viruses, and also increased tolerance of the plants to certain herbicidally active compounds. Examples of transgenic plants which may be mentioned are the important crop plants, such as cereals (wheat, rice), maize, soya beans, 30 potatoes, cotton, tobacco, oilseed rape and also fruit plants (with the fruits apples, pears, citrus fruits and grapes), and particular emphasis is given to maize, soya beans, potatoes, cotton, tobacco and oilseed rape. Traits that are emphasized are in particular increased defence of the plants against insects, arachnids, nematodes and slugs and snails by toxins formed in the plants, in particular those formed in the plants by the genetic material from *Bacillus thuringiensis* (for 35 example by the genes CryIA(a), CryIA(b), CryIA(c), CryIIA, CryIIIA, CryIIIB2, Cry9c, Cry2Ab, Cry3Bb and CryIF and also combinations thereof) (hereinbelow referred to as "Bt plants"). Traits that are also particularly emphasized are the increased defence of the plants against fungi, bacteria and viruses by systemic acquired resistance (SAR), systemin, phytoalexins, elicitors and resistance genes and correspondingly expressed proteins and toxins. Traits that are furthermore particularly

emphasized are the increased tolerance of the plants to certain herbicidally active compounds, for example imidazolinones, sulphonylureas, glyphosate or phosphinotricin (for example the "PAT" gene). The genes which impart the desired traits in question can also be present in combination with one another in the transgenic plants. Examples of "Bt plants" which may be mentioned are 5 maize varieties, cotton varieties, soya bean varieties and potato varieties which are sold under the trade names YIELD GARD® (for example maize, cotton, soya beans), KnockOut® (for example maize), StarLink® (for example maize), Bollgard® (cotton), Nucoton® (cotton) and NewLeaf® (potato). Examples of herbicide-tolerant plants which may be mentioned are maize varieties, cotton 10 varieties and soya bean varieties which are sold under the trade names Roundup Ready® (tolerance to glyphosate, for example maize, cotton, soya bean), Liberty Link® (tolerance to phosphinotricin, for example oilseed rape), IMI® (tolerance to imidazolinones) and STS® (tolerance to sulphonylureas, for example maize). Herbicide-resistant plants (plants bred in a conventional manner for herbicide tolerance) which may be mentioned also include the varieties 15 sold under the name Clearfield® (for example maize). Of course, these statements also apply to plant cultivars which have these genetic traits or genetic traits still to be developed, and which will be developed and/or marketed in the future.

The plants listed can be treated according to the invention in a particularly advantageous manner 20 with the compounds of the general formula (I) or the active compound mixtures according to the invention. The preferred ranges stated above for the active compounds or mixtures also apply to the treatment of these plants. Particular emphasis is given to the treatment of plants with the compounds or mixtures specifically mentioned in the present text.

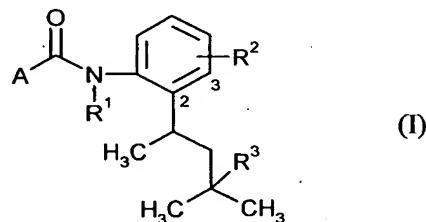
The preparation and the use of the active compounds according to the invention is illustrated by 25 the examples below.

Preparation examplesExample 1

5 191.3 mg (1.0 mmol) of [2-(1,3,3-trimethylbutyl)phenyl]amine are added to a solution comprising 250.2 mg (1.1 mmol) of 3-dichloromethyl-1-methyl-1H-pyrazole-4-carbonyl chloride and 161.9 mg (1.6 mmol) of triethylamine in 10 ml of tetrahydrofuran. The reaction solution is stirred at 60°C for 16 h, filtered through silica and concentrated under reduced pressure.

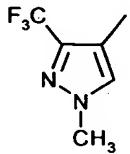
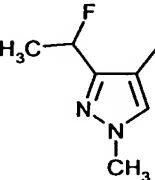
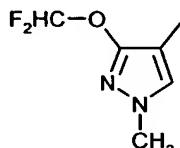
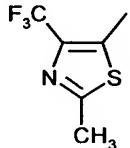
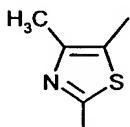
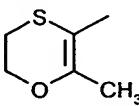
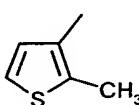
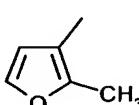
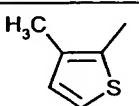
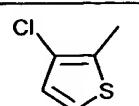
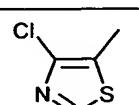
10 Column chromatography (cyclohexane/ethyl acetate 3:1) gives 342.1 mg (89% of theory) of 3-(dichloromethyl)-1-methyl-N-[2-(1,3,3-trimethylbutyl)phenyl]-1H-pyrazole-4-carboxamide [logP (pH = 2.3) = 4.02].

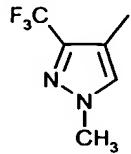
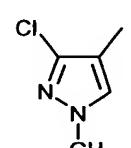
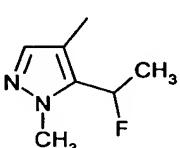
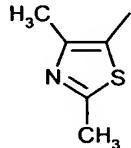
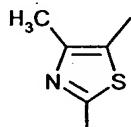
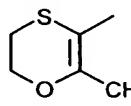
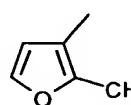
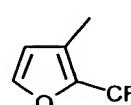
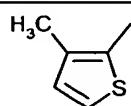
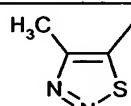
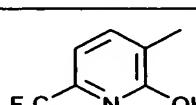
15 The compounds of the formula (I) listed in Table 1 below are obtained analogously to Example 1 and in accordance with the instructions in the general descriptions of the processes.

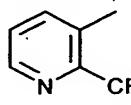
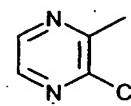
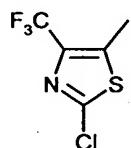
Table 1

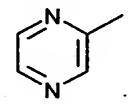
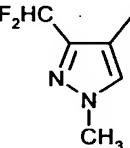
Ex.	R ¹	R ²	R ³	A	logP
2	H	H	CH ₃		3.19
4	H	H	CH ₃		4.25

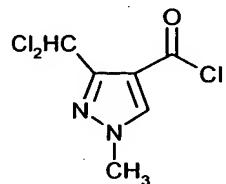
Ex.	R ¹	R ²	R ³	A	logP
3	H	H	CH ₃		4.34
5	H	H	CH ₃		4.39

Ex.	R ¹	R ²	R ³	A	logP
6	H	H	CH ₃		3.81
8	H	H	CH ₃		3.63
10	H	H	CH ₃		4.19
12	H	H	CH ₃		4.24
14	H	H	CH ₃		4.52
16	H	H	CH ₃		4.27
18	H	H	CH ₃		4.39
20	H	H	C ₂ H ₅		4.38
22	H	H	CH ₃		4.40
24	H	H	CH ₃		4.92
26	H	H	CH ₃		4.15

Ex.	R ¹	R ²	R ³	A	logP
7	H	H	C ₂ H ₅		4.13
9	H	H	CH ₃		3.79
11	H	H	CH ₃		3.81
13	H	H	CH ₃		3.60
15	H	H	C ₂ H ₅		4.89
17	H	H	C ₂ H ₅		4.63
19	H	H	CH ₃		4.04
21	H	H	CH ₃		4.37
23	H	H	C ₂ H ₅		4.75
25	H	H	CH ₃		3.84
27	H	H	CH ₃		3.97

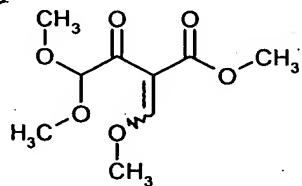
Ex.	R ¹	R ²	R ³	A	logP
28	H	H	CH ₃		3.89
30	H	H	CH ₃		3.95
32	H	H	CH ₃		4.80

Ex.	R ¹	R ²	R ³	A	logP
29	H	H	CH ₃		3.97
31	H	H	CH ₃		4.16

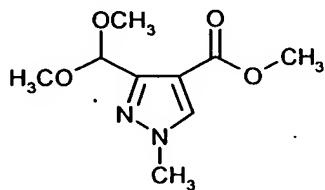
Preparation of starting materials of the formula (II)Example (II-1)

5 300.0 mg (1.9 mmol) of 3-formyl-1-methyl-1H-pyrazole-4-carboxylic acid are dissolved in 60 ml of dichloromethane, and 1.0 g (4.9 mmol) of phosphorus pentachloride is added. After 1.5 h at room temperature, the mixture is poured onto ice-water and extracted with dichloromethane, and the extracts are dried over magnesium sulphate, filtered and concentrated under reduced pressure. This gives 384.0 mg (86% of theory) of 3-dichloromethyl-1-methyl-1H-pyrazole-4-carbonyl chloride

10 [logP (pH 2.3) = 1.80].

Preparation of starting materials of the formula (VII)Example (VII-1)

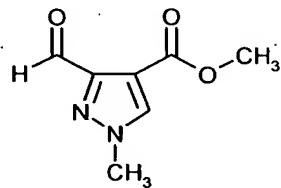
15 16.0 ml (170 mol) of acetic anhydride are added to a solution comprising 10.0 g (57 mmol) of methyl 4,4-dimethoxy-3-oxobutyrate in 9.0 g (85 mmol) of trimethyl orthoformate. The reaction mixture is heated under reflux for 16 h. Distillation of the reaction mixture (boiling point 132-135°C, 0.2 bar) gives 7.0 g (56% of theory) of methyl 4,4-dimethoxy-2-methoxymethylene-3-oxobutyrate.

Preparation of starting materials of the formula (IX)Example (IX-1)

At -5°C , a solution comprising 2.0 ml (38 mmol) of methylhydrazine in 340 ml of methanol is slowly added dropwise to 7.5 g of methyl 4,4-dimethoxy-2-methoxymethylene-3-oxobutyrate. After the addition has ended, the reaction mixture is stirred at room temperature for 16 h and concentrated under reduced pressure.

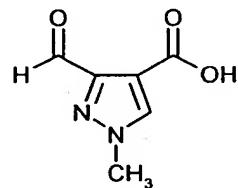
Column chromatography (mobile phase gradient cyclohexane/ethyl acetate) gives 6.5 g (77% of theory) of methyl 3-dimethoxymethyl-1-methyl-1H-pyrazole-4-carboxylate.

10

Preparation of starting materials of the formula (X)Example (X-1)

10 ml of concentrated hydrochloric acid is added to a solution of 2.1 g (10 mmol) of methyl 3-dimethoxymethyl-1-methyl-1H-pyrazole-4-carboxylate in 20 ml of dioxane, and the mixture is stirred at room temperature for 16 h. For work-up, the mixture is concentrated under reduced pressure and the residue is taken up in 200 ml of methylene chloride and washed with 50 ml of water. The organic phase is dried over magnesium sulphate, filtered and concentrated.

This gives 1.6 g (94% of theory) of methyl 3-formyl-1-methyl-1H-pyrazole-4-carboxylate [$\log P$ (pH 2.3) = 0.46].

Preparation of starting materials of the formula (XI)Example (XI-1)

25 6.0 g (35.68 mmol) of methyl 3-formyl-1-methyl-1H-pyrazole-4-carboxylate are dissolved in 180 ml of tetrahydrofuran and 90 ml of water, and 0.94 g (39.25 mmol) of lithium hydroxide is added. The reaction mixture is stirred at room temperature for 16 h, the organic solvent is removed under reduced pressure and the aqueous phase that remains is acidified with dilute hydrochloric acid and extracted

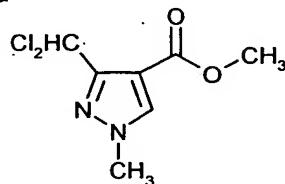
three times with in each case 100 ml of ethyl acetate. The organic phases are dried over magnesium sulphate, filtered and concentrated.

This gives 4.28 g (78% of theory) of 3-formyl-1-methyl-1H-pyrazole-4-carboxylic acid of $\log P$ (pH = 2.3) = -0.19.

5

Preparation of starting materials of the formula (XII)

Example (XII-1)



46.1 mg (0.27 mmol) of methyl 3-formyl-1-methyl-1H-pyrazole-4-carboxylate are dissolved in 10 ml
10 of dichloromethane, and 142.9 mg (0.67 mmol) of phosphorus pentachloride are added. The reaction mixture is stirred at room temperature for 1.5 h, added to water and extracted with diethyl ether, and the extract is dried over magnesium sulphate and concentrated under reduced pressure.

This gives 53.0 mg (86% of theory) of methyl 3-(dichloromethyl)-1-methyl-1H-pyrazole-4-carboxylate of $\log P$ (pH 2.3) = 1.80.

15

This methyl ester can be hydrolysed in the customary manner. This gives 3-(dichloromethyl)-1-methyl-1H-pyrazole-4-carboxylic acid which is coupled with compounds of the formula (III) either directly or after conversion into the acid chloride.

20. The determination of the $\log P$ values given in the preparation examples and tables above is carried out in accordance with EEC Directive 79/831 Annex V.A8 by HPLC (High Performance Liquid Chromatography) on a reversed-phase column (C 18). Temperature: 43°C.

The determination is carried out in the acidic range at pH 2.3 using the mobile phases 0.1% aqueous phosphoric acid and acetonitrile; linear gradient from 10% acetonitrile to 90% acetonitrile.

25. Calibration is carried out using unbranched alkan-2-ones (of 3 to 16 carbon atoms) with known $\log P$ values (determination of the $\log P$ values by the retention times using linear interpolation between two successive alkanones).

The lambda max values were determined in the maxima of the chromatographic signals using the UV spectra from 200 nm to 400 nm.

Use examples

Example A

5 **Podosphaera test (apple) / protective**

Solvents: 24.5 parts by weight of acetone
 24.5 parts by weight of dimethylacetamide
Emulsifier: 1 part by weight of alkylaryl polyglycol ether

10

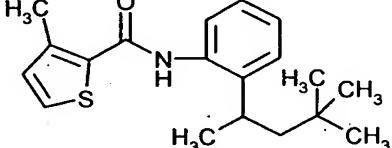
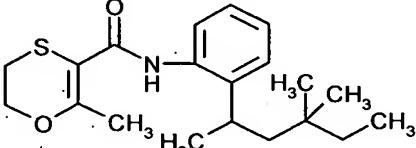
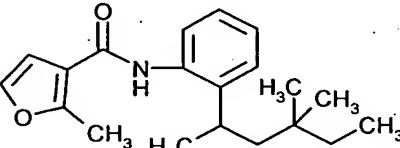
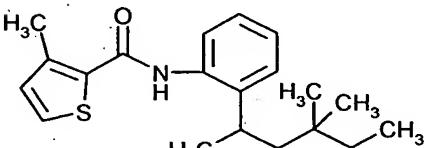
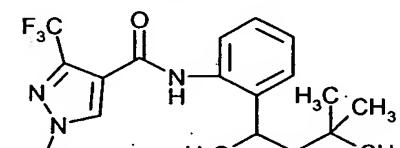
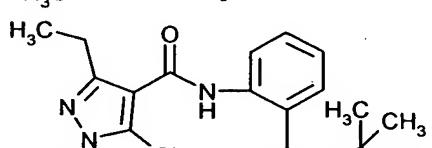
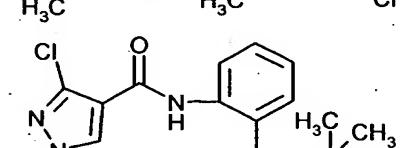
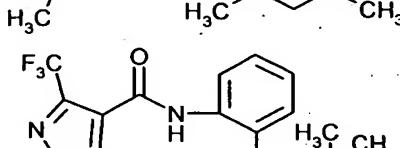
To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

15 To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are inoculated with an aqueous spore suspension of the apple mildew pathogen *Podosphaera leucotricha*. The plants are then placed in a greenhouse at about 23°C and a relative atmospheric humidity of about 70%.

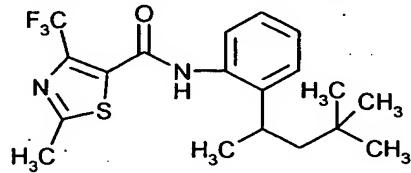
20 Evaluation is carried out 10 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

Table A

Podosphaera test (apple) / protective

Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %
	100	85
	100	94
	100	91
	100	98
	100	95
	100	96
	100	100
	100	97

Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %
---	--	------------------



100

100

Example B

Venturia test (apple) / protective

5 Solvents: 24.5 parts by weight of acetone
24.5 parts by weight of dimethylacetamide
Emulsifier: 1 part by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

15 To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are inoculated with an aqueous conidia suspension of the apple scab pathogen *Venturia inaequalis* and then remain in an incubation cabin at about 20°C and 100% relative atmospheric humidity for 1 day.

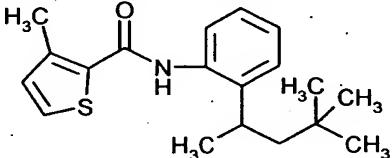
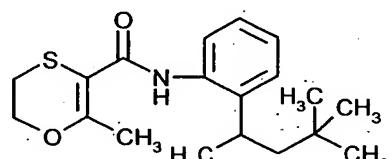
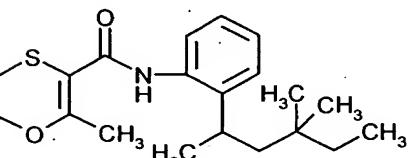
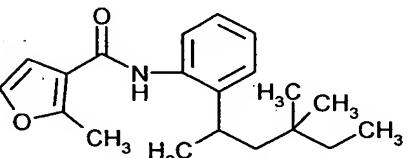
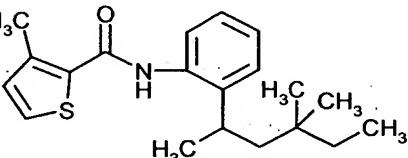
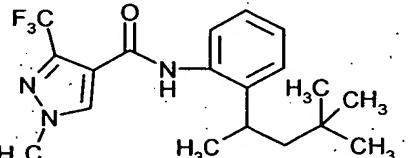
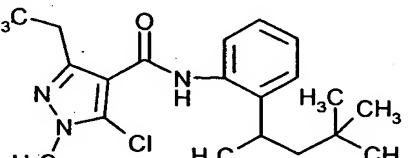
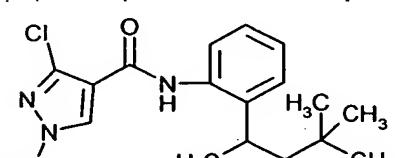
The plants are then placed in a greenhouse at about 21°C and a relative atmospheric humidity of about 90%.

20

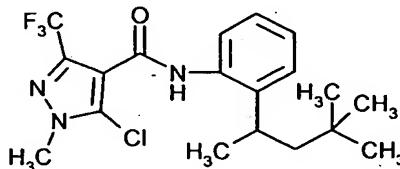
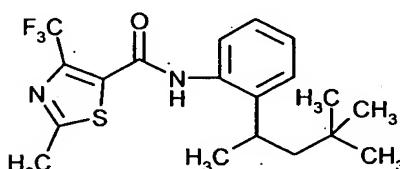
Evaluation is carried out 10 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

Table B

Venturia test (apple) / protective

Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %
	100	96
	100	100
	100	100
	100	93
	100	99
	100	95
	100	92
	100	100

Venturia test (apple) / protective

Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %
	100	99
	100	99

Example C

Botrytis test (bean) / protective

5 Solvents: 24.5 parts by weight of acetone
 24.5 parts by weight of dimethylacetamide
Emulsifier: 1 part by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvents and emulsifier, and the concentrate is diluted with water to the desired concentration.

15 To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, 2 small pieces of agar colonized by Botrytis cinerea are placed onto each leaf. The inoculated plants are placed in a dark chamber at about 20°C and 100% relative atmospheric humidity.

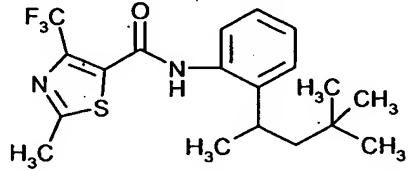
20 2 days after the inoculation, the size of the infected areas on the leaves is evaluated. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

Table C

Botrytis test (bean) / protective

Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %
	500	100
	500	95
	500	100
	500	88
	500	100
	500	100
	500	95
	500	97

Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %
---	--	------------------



500

95

Example D

Puccinia test (wheat) / curative

5 a) Solvent: 50 parts by weight of N,N-dimethylacetamide
 Emulsifier: 1 part by weight of alkylaryl polyglycol ether
b) Solvent: 25 parts by weight of N,N-dimethylacetamide
 Emulsifier: 0.6 part by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

15 To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are sprayed with a conidia suspension of Puccinia recondita. The plants remain in an incubation cabin at 20°C and 100% relative atmospheric humidity for 48 hours.

20 The plants are then placed in a greenhouse at a temperature of about 20°C and a relative atmospheric humidity of 80% to promote the development of rust pustules.

Evaluation is carried out 10 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

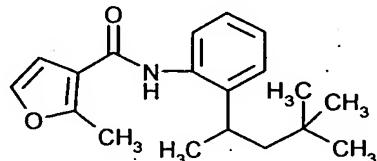
Table D

Puccinia test (wheat) /protective

Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %	Solvent/ emulsifier
	500	100	a)
	500	100	b)

Puccinia test (wheat) /protective

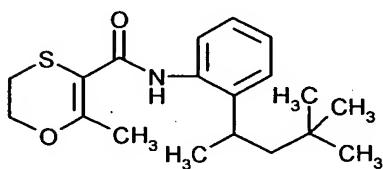
Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %	Solvent/ emulsifier
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500

100

b)



500

100

b)



500

100

b)

Example E

Sphaerotheca test (cucumber) / protective

5 Solvent: 49 parts by weight of N,N-dimethylformamide
Emulsifier: 1 part by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

15 To test for protective activity, young cucumber plants are sprayed with the preparation of active compound at the stated application rate. 1 day after treatment the plants are inoculated with a spore suspension of Sphaerotheca fuliginea. The plants are then placed in a greenhouse at a relative atmospheric humidity of 70% and a temperature of 23°C.

Evaluation is carried out 7 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

Table E

Sphaeroteca test (cucumber) / protective

Active compound According to the invention	Application rate of active compound in g/ha	Efficacy in %
	750	95
	750	95
	750	90

Example F

Erysiphe test (barley) / protective

5 Solvent: 25 parts by weight of N,N-dimethylacetamide
Emulsifier: 0.6 part by weight of alkylaryl polyglycol ether

10 To produce a suitable preparation of active compound, 1 part by weight of active compound is mixed with the stated amounts of solvent and emulsifier, and the concentrate is diluted with water to the desired concentration.

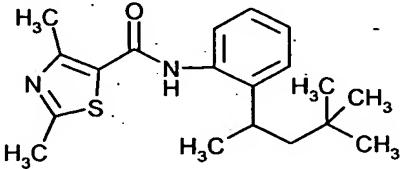
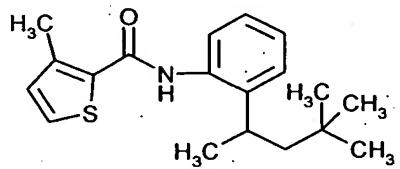
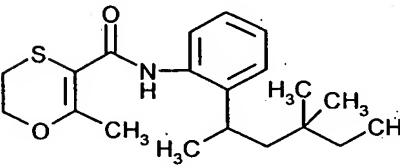
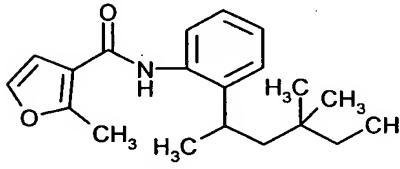
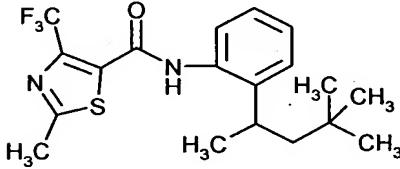
To test for protective activity, young plants are sprayed with the preparation of active compound at the stated application rate. After the spray coating has dried on, the plants are dusted with spores of *Erysiphe graminis* f.sp. *hordei*.

15 The plants are placed in a greenhouse at a temperature of about 20°C and a relative atmospheric humidity of about 80% to promote the development of mildew pustules.

20 Evaluation is carried out 7 days after the inoculation. 0% means an efficacy which corresponds to that of the control, whereas an efficacy of 100% means that no infection is observed.

Table F

Erysiphe test (barley) / protective

Active compound According to the invention.	Application rate of active compound in g/ha	Efficacy in %
	500	100
	500	100
	500	94
	500	100
	500	100